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DEPARTMENT OF COMMERCE
BUREAU OF STANDARDS

S. W. STRATTON, Director

DENSITY AND THERMAL EXPANSION OF ETHYL
ALCOHOL AND OF ITS MIXTURES WITH WATER

BY

N. S. OSBORNE, Associate Physicist

E. C. McKELVY, Assistant Chemist

and

H. W. BEARCE, Assistant Physicist

Bureau of Standards

[APRIL 15, 1913]

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By N. S. Osborne, E. C. McKelvy, and H. W. Bearce

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INTRODUCTION

The demand for alcoholometric density tables of greater precision than those now in use has led to the experimental re-determination at the Bureau of Standards of the constants upon which such tables are based.

In order to provide the basis for the standardization of alcoholometers at this bureau, the work of preparing such tables was undertaken several years ago. Examination of the data then available showed that they were inadequate for the preparation of tables of the desired accuracy.

The scope of the present work includes the preparation of pure anhydrous ethyl alcohol, the determination of the density and thermal expansion of this alcohol and its mixtures with water, and the construction from these data of tables for convenient use.

The results of the density determinations by the authors are expressed in terms of the density of water at 4° C as the unit. The word "density" is employed throughout the paper in a general way as the name of the property of the substance; but where numerical values are given, whether those determined by the

authors or quoted from other observers, the unit employed is either expressed in words or indicated by symbols.

The symbol $D_{\overline{T}}^t$ indicates the density of a substance at the temperature t in terms of the density of water under normal atmospheric pressure at the temperature T as the unit. The density expressed thus is equivalent to the ratio ^a of the density of the substance at t° to the density of water at T° , and in the case where T equals 4° C the density is equivalent to density expressed in grams per millimeter. If not otherwise indicated, temperatures are on the centigrade scale, and for the experimental work presented here the International Hydrogen Scale of temperatures is used.

^a This ratio is often called "specific gravity."

PART 1

PREPARATION OF PURE ANHYDROUS ETHYL ALCOHOL

By E. C. McKelvy

The success of an attempt to prepare a pure material of any kind depends largely on its physical state at ordinary temperatures and on the limitations set by the nature of the physical properties, which may be determined in order to detect the impurities present. Generally speaking, chemical methods for detecting very small quantities of one substance in large quantities of another are rather poorly developed and unsatisfactory, especially in the field of organic substances. The present work presented the problem of preparing a pure liquid, ethyl alcohol, with many favorable conditions under which to work. The impurities contained in the product on the market are comparatively few, and there is no great difficulty in getting large quantities of the liquid containing 99.5 per cent ethyl alcohol. The problem resolved itself into a removal of the half per cent of impurity, mostly water, and the study of the variation of the physical properties of alcohol with small amounts of various impurities. The purification can be carried out by distillation at temperatures easily attainable, and the permissibility of working with large quantities of the liquid is a distinct advantage, especially in the preliminary purification. A considerable number of physical properties are applicable to the testing of the product, with, however, a large variation in their sensitiveness. It is evident that our knowledge of the purity of any material is subject to the limits set by the method of testing, whatever may be the efficiency of the method of purification.

I. HISTORICAL SURVEY

It was early appreciated that the most convenient basis upon which alcoholic liquors could be taxed was their alcohol content. The accurate determination of this value has occupied the attention of investigators for over a century, and several independent

methods have been devised. A study of the literature on the subject will show that a number of physical constants have been proposed for this use. By far the larger bulk of the work, however, has had to do with the determination of the densities of alcohol-water mixtures, since a density determination gives an easy and accurate method of determining the alcoholic content of a liquor.

To utilize this method practically, it is of course necessary first to construct a table from density determinations on mixtures of known composition. In order that this may be done, it is essential that pure alcohol should be obtained. The efforts at the preparation of such an alcohol have been parallel with the development in the accuracy of alcoholometric tables. It is purposed to give a short historical survey of the main attempts to accomplish this result.

1. WORK BEFORE 1865—COMPARISON OF RESULTS.

It can be said concerning most of the results recorded before 1865, and of many since, that they are of limited accuracy, and hence of interest mainly from a historical point of view, notwithstanding that a part of this work forms the basis of the tables in current use. The main cause of this status is that the actual density determinations are generally given only to the fourth decimal place, and as a rule the results are poorly defined as to temperature conditions and bases of temperature and density references. A considerable part of this work has been briefly sketched in the papers of Mendeléeff ^{b, 207, 241} and Gerlach ⁴⁴⁵. Blagden and Gilpin ^{4, 5} were among the first investigators to construct tables from actual density determinations. Their work was incomplete in that no success attended their efforts to obtain anhydrous alcohol, and so their results had only an arbitrary significance until the real alcohol value of their basis could be determined by later observers. The early attempts of Lowitz ¹⁰ and Richter ^{9, 11} to dehydrate alcohol, and the work of Tralles ²⁰ have been thoroughly discussed by Windisch ⁶⁸⁴. Table I will show the surprising agreement shown in the results of some of

^b Reference numbers printed in superior figures throughout this paper refer to "Bibliography," part 5, in which each title bears a specific reference number.

the experimenters working before 1865. McCulloch ^{105, 119} in a report to the Secretary of the Treasury of the United States reviews the work done up to that time, and on the basis of a few experiments and recalculations of his own recommends the alcoholometric tables for official use by the Treasury Department.

TABLE I
Density Determinations of Absolute Alcohol Prior to 1866

Name	Year	Dehydrator	Density value $D_{\frac{t}{T}}$	Basis of reference $\frac{t}{T}$	Density $\frac{20^{\circ}}{4^{\circ}}$ recalculated	Remarks
Blagden and Gilpin ^{4, 5}	{ 1790 to 1794 }	None.....	0.825	$\frac{60^{\circ}}{60^{\circ}}F$	{ No dehydrating agent
Lowitz ¹⁰	1796	K ₂ CO ₃791	$\frac{16^{\circ}}{16^{\circ}}C$	0.7899	M.
Richter ^{9, 11}	1797	CaCl ₂792	$\frac{20^{\circ}}{20^{\circ}}C$.7909	M.
Saussure ¹⁷	1807	CaCl ₂792	$\frac{16^{\circ}}{16^{\circ}}C$.7909	M.
Gay-Lussac ²⁶	1815	CaO.....	.7940	$\frac{15^{\circ}}{4^{\circ}}C$.7898	M.
Meissner ²⁷	1816	CaCl ₂791	$\frac{20^{\circ}}{20^{\circ}}C$.7899	M.
Delezennes ³²	182379370	$\frac{20^{\circ}}{0^{\circ}}C$.79361	M.
Gouvernain ³³	182579364	$\frac{16^{\circ}}{10^{\circ}}R$.79348	M.
Dumas and Boullay ⁴¹	1827	CaCl ₂7915	$\frac{18^{\circ}}{X^{\circ}}C$.7899	A.
Muncke and Gmelin ^{47, 52} ..	18288062	$\frac{0^{\circ}}{4^{\circ}}C$.7895	M.
Brown ⁴⁸	1829791
Connell ⁵³	1835	CaO.....	.7938	$\frac{60^{\circ}}{X^{\circ}}F$.7896	A. Assume $x^{\circ}=60^{\circ}F$
Kopp ⁸³	184580950	$\frac{0^{\circ}}{0^{\circ}}C$.79277	M.
Pierre ⁸⁴	1845	CaO.....	.81508	$\frac{0^{\circ}}{4^{\circ}}C$.79777	M. Pierre's expansion coefficient used
Fownes ⁹⁰	1847	CaO.....	.7938	$\frac{60^{\circ}}{60^{\circ}}F$.78959	M.
Wackenroder ⁹⁷	1847	CaO.....	.7948	$\frac{21^{\circ}}{X^{\circ}}C$.7905	A.

TABLE I—Continued

Name	Year	Dehydrator	Density value $D_{\frac{t}{T}}$	Basis of reference $\frac{t}{T}$	Density $\frac{20^{\circ}}{4^{\circ}}$ recalculated	Remarks
Drinkwater ¹⁰²	1848	CaO.....	0.793811	$\frac{60^{\circ}}{60^{\circ}}\text{F}$	0.78958	M.
Wetherill ¹⁰⁹	18488194	$\frac{19^{\circ}}{X^{\circ}}\text{C}$.8073	A.
Plücker ¹³²	1854	CaO.....	.792	$\frac{13^{\circ}75}{X^{\circ}}\text{C}$.787	A.
Delffs ¹²⁸	1854809	$\frac{5^{\circ}}{X^{\circ}}\text{C}$.796	A.
Pouillet ¹⁵²	18597947	$\frac{15^{\circ}}{15^{\circ}}\text{C}$.7898	M.
Baumhauer ¹⁵⁵	1860	CaO.....	.7939	$\frac{15^{\circ}}{4^{\circ}}\text{C}$.7897	A.
Mendeléeff ¹⁷²	18617958	$\frac{15^{\circ}}{4^{\circ}}\text{C}$.7916	A.
Gladstone-Dale ¹⁸⁸	18637972	$\frac{20^{\circ}}{X^{\circ}}\text{C}$.7929	A.
Landolt ¹⁹⁹	1864	CaO-Na....	.8011	$\frac{20^{\circ}}{20^{\circ}}\text{C}$.7997	A.
Mendeléeff ^{207, 241}	1865	CaO-BaO...	.78945	$\frac{20^{\circ}}{4^{\circ}}\text{C}$.78945	Uncorrected

EXPLANATION.—In the column headed “Remarks” M indicates that the densities at 20° were calculated by Mendeléeff. A indicates that this was done by the authors of this paper. Both calculations were made, except where otherwise stated, with Kopp’s ^{92, 241} value for the expansion in the following formula:

$$D_{\frac{t}{4^{\circ}}}^{\frac{t}{4^{\circ}}} = D_{\frac{20^{\circ}}{4^{\circ}}} + (20^{\circ} - t^{\circ})0.000846$$

where 0.000846 is the change in density for 1° C. X° signifies that the basis of density reference was not given and is assumed to be 4° in the recalculation. It is impossible to correct any of these values, except Mendeléeff’s ^{207, 241} later one, to the international hydrogen scale of temperature.

2. WORK OF MENDELÉEFF (1865)

In 1865 Mendeléeff's ²⁰⁷ memoir on alcohol appeared in the original Russian and also in German. His experimental work is characterized by careful attention to details and skillful execution. The ethyl alcohol prepared by him was probably the purest prepared up to that time. His density values have been generally regarded with great confidence. Many of the generally accepted tables used in scientific work have as their basis Mendeléeff's value, with the application of certain well-defined corrections. Such are the tables prepared by Windisch,⁶⁸⁵ using the value approved by the Normal-Eichungs Commission, Berlin; the tables given by E. W. Morley,¹⁰⁵⁰ using a value recalculated by himself; and the tables issued provisionally by the Bureau of Standards ¹²¹⁶ in 1909.

In Mendeléeff's work the first attempt was made to determine the relative efficiency of different dehydrating agents. From the results of previous investigators, as well as experiments of his own, he decided that such agents as anhydrous potassium carbonate, calcium chloride, and copper sulphate were unsatisfactory. Metallic sodium as such or in the form of an amalgam he found inefficient, an observation later substantiated by Squibb ⁴³³ and by Crismer.¹⁰³³ He concluded that lime, with the addition of a smaller quantity of baryta, was the most satisfactory means for dehydrating the alcohol. This addition of baryta had been recommended by Berthelot,¹⁴¹ with the statement that the turning brown of the alcohol was an indication of complete dehydration, but it was later shown by Crismer ¹⁰³³ that the baryta is unnecessary and undesirable.

In Table II are given the lowest density values found by Mendeléeff in his work with different dehydrating agents, certain of which are still in use. These results, if more generally known, would leave no doubt in the minds of chemists as to the agents which should be used if an alcohol as anhydrous as possible is desired. Table III contains the results of one of Mendeléeff's and one of Squibb's later distillations, showing the variation in the density of the different fractions. Unfortunately the volumes of the different fractions are not given.

TABLE II

Densities with Various Dehydrating Agents (Mendeléeff ^{207,241})

Dehydrator used	$D_{\frac{20^{\circ}}{4^{\circ}}}$	Remarks
K ₂ CO ₃	0.78970	All distillations were made with the dehydrating agent present in the alcohol. No corrections have been applied to the results.
CaCl ₂	.78960	
CuSO ₄	.78961	
Na-Hg	.78974	
BaO	.789453	
CaO+BaO	.78945	

TABLE III

Variations in Density of Distillate Fractions

[Dehydrating agents: Mendeléeff, CaO+BaO; Squibb, CaO.]

Fraction	$D_{\frac{20^{\circ}}{4^{\circ}}}$ Mendeléeff ^{207, 241}	$D_{\frac{15.96}{15.96}}$ E. R. Squibb ⁶⁷³	Remarks
1	0.78963	0.793960	These results are taken directly from the original articles without correction. The volume figures for the fractions are not available.
2	.78946	.793811	
3	.789442	.793639	
4	.789456	.793582	
5	.789442	.793576	
6	.789472	.793561	
7793499	

3. WORK SUBSEQUENT TO 1865

In Table IV are presented the results of most importance which have appeared since 1865. Table V shows the values obtained by other investigators during this period in cases where generally the density value was of secondary importance to the determination of other physical constants. In most of these cases no details of the density determinations are given.

TABLE IV

Ethyl Alcohol Density Determinations of Most Importance Subsequent to 1864

Name	Year	Dehydrator used	Density value given $D_{\frac{t}{T}}$	Basis of reference $\frac{t}{T}$	Density at 25° (cal- culated) $D_{\frac{25}{4}}^C$
Mendeléeff ^{207,241}	1865	CaO+BaO.....	0. 79358	$\frac{15^{\circ}}{4^{\circ}}$	0. 78504
Squibb, Messrs. ⁴³³ ..	1884	CaO.....	. 79350	$\frac{15^{\circ}6}{15^{\circ}6}$. 78470
Squibb, E. R. ⁶⁷⁹	1893	CaO.....	. 79356	$\frac{15^{\circ}6}{15^{\circ}6}$. 78476
Cook and Haines ⁹³³ .	1901	CaC ₂ 79357	$\frac{15^{\circ}}{4^{\circ}}$. 78503
Young ⁹⁸⁶	1902	Dist. w. hexane.....	. 80627	$\frac{0^{\circ}}{4^{\circ}}$. 78507
Crismer ¹⁰³³	1904	CaO.....	. 78746	$\frac{25^{\circ}}{25^{\circ}}$. 78516
Winkler ¹⁰⁸⁷	1905	Ca.....	. 78932	$\frac{20^{\circ}07}{4^{\circ}}$. 78510
Klason and Norlin ¹¹⁰⁹	1906	Ca.....	. 79413	$\frac{15^{\circ}}{15^{\circ}}$. 78490
Do.....		Ca.....	. 78938	$\frac{20^{\circ}}{4^{\circ}}$. 78508
Kailan ¹¹⁴⁸	1907	CaO.....	. 78520	$\frac{25^{\circ}}{4^{\circ}}$. 78520
Andrews ¹¹⁶⁷	1908	CaO, Ca, Mg-Hg...	. 78510	$\frac{25^{\circ}}{4^{\circ}}$. 78510
Doroshevskii ¹¹⁷⁵	1908	CaO.....	. 79426	$\frac{15^{\circ}}{15^{\circ}}$. 78503
Acree and Robert- son ^{1257, 1294}	1910	CaO.....	. 78506	$\frac{25^{\circ}}{4^{\circ}}$. 78506
Doroshevskii ¹²⁶⁹	1910	CaO.....	. 79412	$\frac{15^{\circ}}{15^{\circ}}$. 78489
Röhrs ¹²⁹⁵	1910	Ca.....	. 795027	$\frac{14^{\circ}2}{14^{\circ}2}$. 78521
Kailan ¹³⁴¹	1911	CaO.....	. 78513	$\frac{25^{\circ}}{4^{\circ}}$. 78513
Bureau Standards ¹³⁷⁶	1910	CaO, Al-Hg.....	. 78506	$\frac{25^{\circ}}{4^{\circ}}$. 78506

EXPLANATION.—Table IV represents an attempt to put the most important results for the density of anhydrous alcohol on a comparable basis. All reductions were made with the use of the formula obtained in Part II,

$$D_{4}^{t^{\circ}} = D_{4}^{25^{\circ}} - [859 (t - 25) + 0.6 (t - 25)^2 + 0.005 (t - 25)^3] \times 10^{-6}$$

and Chappuis's ¹¹³² values for the density of water. The value given in Mendeléeff's original paper has been corrected to a more probable value for the density of water than that assumed by him and to the international hydrogen temperature scale.

TABLE V

Density Determinations of Secondary Importance Subsequent to 1865

Name	Year	Dehydrator	Density value $D_{\overline{T}}^t$	Basis of reference $\frac{t}{\overline{T}}$	Density at 25° (calculated) $D_{\overline{4}}^{25}$
Linnemann ²³¹	1868	K ₂ CO ₃	0. 8086	$\frac{19^{\circ}}{19^{\circ}}$	0. 8022
Wüllner ²³⁴	1868 8133	$\frac{0^{\circ}}{x^{\circ}}$. 7920
Darling ²²⁵	1868 8095	$\frac{0^{\circ}}{x^{\circ}}$. 7926
Dupré and Page ²³⁷	1869 79792	$\frac{10^{\circ}}{x^{\circ}}$. 78516
Do.....	1869 79317	$\frac{15^{\circ}5}{x^{\circ}}$. 78509
Do.....	1869 78932	$\frac{20^{\circ}}{x^{\circ}}$. 78507
van der Willigen ²⁴⁵	1869 7935	$\frac{20^{\circ}}{4^{\circ}}$. 7892
Pierre and Puchot ²⁶⁰	1871 822	$\frac{20^{\circ}}{x^{\circ}}$. 818
Erlenmeyer ²⁵⁴	1871	CaO.....	. 79481	$\frac{11^{\circ}}{x^{\circ}}$. 78291
Pierre ²⁸³	1873 80214	$\frac{15^{\circ}}{x^{\circ}}$. 79460
Winkelmann ²⁸⁵	1873 7946	$\frac{16^{\circ}03}{4^{\circ}}$. 78695

TABLE V—Continued

Name	Year	Dehydrator	Density value $D_{\frac{t}{T}}$	Basis of reference $\frac{t}{T}$	Density at 25° (calculated) $D_{\frac{25}{4}}$
Duclaux ³¹²	1877	0. 7947	$\frac{15^{\circ}}{4^{\circ}}$	0. 7860
Kundt ³²²	1878 800	$\frac{18^{\circ}}{x^{\circ}}$. 794
Brühl ³⁴³	1880 8000	$\frac{20^{\circ}}{4^{\circ}}$. 7957
De Heen ³⁴⁷	1880 7995	$\frac{14^{\circ}}{x^{\circ}}$. 7901
Lorenz ³⁵⁰	1880 7909	$\frac{20^{\circ}}{x^{\circ}}$. 7866
Vincent and Delachanal ³⁵⁹ ..	1880 8120	$\frac{0^{\circ}}{x^{\circ}}$. 7908
Zettermann ³⁶²	1880 799	$\frac{17^{\circ}5}{x^{\circ}}$. 793
Bedson and Williams ³⁶⁴	1881 8019	$\frac{20^{\circ}}{4^{\circ}}$. 7976
Drecker ⁴⁰⁰	1883 78962	$\frac{25^{\circ}42}{x^{\circ}}$. 78998
Johst ⁴⁰⁵	1883 8072	$\frac{16^{\circ}7}{4^{\circ}}$. 8000
Nasini ⁴⁰⁸	1883	BaO.....	. 7968	$\frac{20^{\circ}}{4^{\circ}}$. 7925
Quincke ⁴¹¹	1883 7969	$\frac{20^{\circ}}{4^{\circ}}$. 7926
Sieben ⁴¹⁴	1883 796	$\frac{20^{\circ}}{4^{\circ}}$. 792
Perkin ⁴³¹	1884	CuSO ₄ 78820	$\frac{25^{\circ}}{25^{\circ}}$. 78589
Kanonnikoff ⁴⁴⁹	1885 7918	$\frac{20^{\circ}}{4^{\circ}}$. 7875
Winkelmann. ⁴⁶² (Kahl- baum's alcohol.)	1885 7906	$\frac{20^{\circ}}{4^{\circ}}$. 7863

TABLE V—Continued

Name	Year	Dehydrator	Density value $D_{\frac{t}{T}}$	Basis of reference $\frac{t}{T}$	Density at 25° (calculated) $D_{\frac{25}{4}}$
Pagliani and Batelli ⁴⁵³	1885	0. 79175	$\frac{18^{\circ}}{x^{\circ}}$	0. 78580
Worthington ⁴⁶³	1885 7906	$\frac{25^{\circ}3}{25^{\circ}}$. 7885
Ketteler ⁵³¹	1388 78987	$\frac{20^{\circ}}{x^{\circ}}$. 78562
Do.....	1888 80681	$\frac{0^{\circ}}{x^{\circ}}$. 78560
Ångstrom. ⁵¹² (Air free).....	1888 80715	$\frac{0^{\circ}}{4^{\circ}}$. 78595
Ångstrom. (Air saturated)...	1888 80680	$\frac{0^{\circ}}{4^{\circ}}$. 78559
Hartwig ⁵²⁷	1888 7937	$\frac{18^{\circ}}{4^{\circ}}$. 7877
Le Blanc ⁵⁶³	1889 79643	$\frac{20^{\circ}}{20^{\circ}}$. 79076
Barbier and Roux ⁵⁷³	1890 805	$\frac{14^{\circ}}{x^{\circ}}$. 796
Buchkremer ⁵⁸¹	1890 7935	$\frac{20^{\circ}}{4^{\circ}}$. 7892
Gartenmeister ⁵⁸⁴	1890 7943	$\frac{20^{\circ}}{4^{\circ}}$. 7900
Korten ⁵⁸⁸	1890 7910	$\frac{20^{\circ}}{4^{\circ}}$. 7868
Jahn ⁶⁰⁹	1891 79149	$\frac{20^{\circ}}{20^{\circ}}$. 78585
Schall and Koss'ky ⁶²²	1891 791861	$\frac{18^{\circ}}{4^{\circ}}$. 785883
Landolt and Jahn ⁶⁴⁶	1892 80197	$\frac{17^{\circ}5}{x^{\circ}}$. 79557
Eykmann ⁶⁵⁹	1893 7963	$\frac{12^{\circ}7}{4^{\circ}}$. 7856

TABLE V—Continued

Name	Year	Dehydrator	Density value $D_{\frac{t}{T}}$	Basis of reference $\frac{t}{T}$	Density at 25° (calculated) $D_{\frac{25}{4}}$
Edwards ⁶⁹²	1894	0. 7964	$\frac{20^{\circ}}{15^{\circ}.5}$	0. 7914
Tammann and Hirsch'g ⁷¹⁷ ..	1894 7951	$\frac{15^{\circ}}{4^{\circ}}$. 7866
Lehfeldt ⁷⁴¹	1895	CaO–BaO 7929	$\frac{18^{\circ}}{4^{\circ}}$. 7869
Sohet ⁸¹⁸	1897 8089	$\frac{0^{\circ}.9}{4^{\circ}}$. 7885
Do.....	1897 7711	$\frac{44^{\circ}.7}{4^{\circ}}$. 7879
Zecchini ⁸³²	1897 80513	$\frac{1^{\circ}.8}{4^{\circ}}$. 78546
Cohen ⁸³⁷	1898	CaO.....	. 8063	$\frac{0^{\circ}}{x^{\circ}}$. 7851
Tanatar and Klimenko ⁸⁶⁰ ...	1898 79565	$\frac{15^{\circ}}{x^{\circ}}$. 78711
Loomis (Squibb). ⁹²⁰ (Deter- mined by E. R. Squibb.)	1900	CaO.....	. 79386	$\frac{15^{\circ}.6}{15^{\circ}.6}$. 78506
Carrara and Levi ⁹⁵⁹	1902 79425	$\frac{16^{\circ}}{4^{\circ}}$. 78741
Grunmach ⁹⁶⁶	1902 7888	$\frac{22^{\circ}.8}{4^{\circ}}$. 7862
Szilard ¹⁰⁸³	1905	CaO.....	. 78990	$\frac{20^{\circ}}{x^{\circ}}$. 78565
Cheneveau ¹¹³³	1907 7886	$\frac{22^{\circ}}{4^{\circ}}$. 7860
Timmermanns ¹¹⁶²	1907 80664	$\frac{0^{\circ}}{4^{\circ}}$. 78544
Wagner and Schulze. ¹¹⁶⁴ (Alcohol from Winkler.)	1907	Ca.....	. 793405	$\frac{15^{\circ}}{4^{\circ}}$. 78486
Getman ¹¹⁸³	1908 78684	$\frac{25^{\circ}}{4^{\circ}}$. 78684

TABLE V—Continued

Name	Year	Dehydrator	Density value $D_{\frac{t}{T}}$	Basis of reference $\frac{t}{T}$	Density at 25° (calculated) $D_{\frac{25}{4}}$
Herz and Kuhn ¹¹⁸⁷	1908	0. 7867	$\frac{25^{\circ}}{4^{\circ}}$	0. 7867
Richards and Mathews. ¹¹⁹⁷ (95 per cent probably.)	1908 8040	$\frac{20^{\circ}}{4^{\circ}}$. 7997
Rühlemann ¹²⁰⁰	1908 7927	$\frac{20^{\circ}}{4^{\circ}}$. 7884
Turner ¹²⁰⁸	1908 78948	$\frac{20^{\circ}}{4^{\circ}}$. 78520
Holmes and Sageman ¹²³² ...	1909	CaO.....	. 78892	$\frac{25^{\circ}}{25^{\circ}}$. 78661
Dawson ¹²⁶⁶	1910 791502	$\frac{18^{\circ}}{4^{\circ}}$. 78552
Polowzow ¹²⁹²	1910 78970	$\frac{20^{\circ}}{4^{\circ}}$. 78542
Smits and de Leeuw ¹³⁰⁸	1910 7907	$\frac{18^{\circ}}{4^{\circ}}$. 7847
Thole ¹³⁰⁹	1910	Ca.....	. 7876	$\frac{25^{\circ}}{4^{\circ}}$. 7876
Warren ¹³²²	1910	CaO.....	. 7940	$\frac{15^{\circ}}{15^{\circ}}$. 7848
Cederberg ¹³³⁰	1911 7907	$\frac{19^{\circ}.5}{x^{\circ}}$. 7860

The density value obtained by Messrs. Squibb ⁴³³ is the lowest ever obtained. These investigators used freshly ignited lime as dehydrating agent. The treatment occupied several weeks and was made by percolation at ordinary temperatures. This work repeated by E. R. Squibb ⁶⁷⁹ 10 years later showed results only slightly higher. These authors concluded that they had not even then obtained anhydrous alcohol. The variation in density of their alcohol from that of Mendeléeff would represent an impurity of 0.03 per cent water in Mendeléeff's alcohol, assuming

his higher value to be due to the presence of water. In the light of data to be presented later, it seems that these results can be explained on no other grounds than that in some way not known their standards of reference differed from those of other observers. In addition, there is some doubt concerning the precision of their temperature measurements. Cook and Haines⁹³³ neglected to give their basis of density reference. On the assumption that this is water at its maximum density it is found to differ only slightly from the more reliable results. This is the only quantitative result obtained, so far as known, by the use of calcium carbide as dehydrating agent. Young,⁹⁸⁶ without the use of chemical agents in the ordinary sense, obtained results in good accord with those of Mendeléeff. This was accomplished by distilling with benzene and with hexane. Crismer¹⁰³³ has shown the inefficiency of sodium and baryta as dehydrating agents and obtained his best alcohol by means of dehydration with lime. He also introduced the use of the critical solution temperature of alcohol-kerosene mixtures as a criterion of the water content of the alcohol. He concluded that its sensibility was of the same order as density determinations accurate to the fifth decimal place. Vandam¹¹²⁰ in Holland and Andrews¹¹⁶⁷ in this country have also made use of this criterion. The use of this constant promises to be of considerable importance both from the standpoint of convenience and range of applicability in determining the composition of water-alcohol mixtures.

Winkler¹⁰⁸⁷ obtained an alcohol of low density, using calcium to remove the water. Klason and Norlin,¹¹⁰⁹ using the same agent after purifying by recrystallization of potassium ethyl sulphate according to recalculation in Table IV, get one result considerably lower than that of Winkler,¹⁰⁸⁷ or even the recalculated result of Mendeléeff.²⁰⁷ However, the authors state that their results confirm the Windisch⁶⁸⁵ basis, which is recalculated from the results of Mendeléeff, and also confirm the result of Winkler¹⁰⁸⁷ as opposed to that of Mendeléeff. These statements can be reconciled with each other and with the results gotten by recalculation, using the thermal expansion formula obtained in part 2, only by the consideration that the authors did not con-

sider 0.05 per cent water, corresponding to 16 units in the fifth decimal place, as significant. The concordance between Mendeléeff's corrected value and Winkler's value is much better than between the latter and the result of Klason and Norlin. The result given under density $\frac{20^\circ}{4^\circ}$ calculated to 25° leaves nothing to be desired when compared with the results of most other investigators. There is some possibility of a misprint in the value at $\frac{15^\circ}{15^\circ}$. Kailan^{1148, 1341} determined the conditions under which lime could be most effectively used as dehydrating agent, and later tried metallic calcium for removing the last traces of water. Andrews¹¹⁶⁷ tested the relative effectiveness of lime, metallic calcium, and magnesium amalgam as dehydrators and obtained alcohols with each that differed only slightly among themselves and were only slightly higher in density than Mendeléeff's²⁰⁷ generally accepted value. He used in addition to the density determinations the critical solution temperature and refractive index as criteria of complete dehydration. Extensive work has been done recently in Russia by Doroshevskii^{1175, 1269} and collaborators on the physical constants of anhydrous alcohol and its mixtures with water. The density value shown in the table was gotten by dehydrating with lime. Among the physical constants considered were specific heat, refractive index, electrical conductivity, vapor pressure, and boiling points. Other alcohols were also studied. The difference in the two density values given for anhydrous ethyl alcohol by this investigator at different times represents 0.04 per cent water.

The determination of the influence of small quantities of water in ethyl alcohol upon the velocity of esterification by H. Goldschmidt and E. Sunde,¹¹⁰¹ and upon the velocity of other reactions by G. Bredig and W. Fraenkel,^{1062, 1090} gives some promise of being of value in estimating the small quantities of water. J. Gyr¹¹⁸⁵ has used this influence as a criterion in the dehydration of methyl alcohol. Millar¹²⁸⁸ and Braune¹³²⁹ regard this method as the best for detecting traces of water in alcohol. In the case of methyl alcohol and the esterification of phenyl acetic acid, assuming the same accuracy of temperature control, the greater accuracy of

the density determination seems to just about counterbalance the great change in esterification constant for the same percentage of water content. However, the quantitative results are as yet too meager to allow of a certain conclusion in regard to the use of this constant. Kailan¹³⁴¹ in a recent work shows the superiority of the density method in one special case. Acree¹²⁵⁷ and students, in velocity of reaction studies, have used an alcohol showing the same density as that obtained at this bureau. The effect of small quantities of water was small in alkaline solution but very marked in acid solution.

II. EXPERIMENTAL PART

1. OUTLINE OF WORK

This part of the work has to do with the preparation of chemically pure ethyl alcohol. Experimentally it was proposed: First, to obtain, if possible, using different methods of purification and dehydration, an alcohol showing constant physical properties, even though from different commercial sources; second, to use density determinations as the main criterion of the degree of dehydration and purity with the auxiliary use, in some cases, of the critical solution temperature of its mixtures with kerosene and of such chemical tests as can be applied; third, to obtain, if possible, an alcohol which on further treatment with dehydrating agents would show no decrease in density and no variation in the density of the various distillates from the beginning to the end of a distillation; fourth, to investigate the possible effect on the density of the impurities likely to occur. Aside from considerations having to do with alcoholometry, it was hoped that the additional information gained regarding the most efficient method for preparing pure alcohol, which as a solvent ranks next to water in importance, would be of some importance and value.

Full details of the density determinations are given on page 405 and following.

2. DETERMINATION OF THE CRITICAL SOLUTION TEMPERATURE

Alexejew,⁴⁶⁴ Rothmund,⁸⁵⁹ and Timmermans¹¹¹⁶ have worked extensively with the mutual solubility relations of two liquids. Consider curve III, Fig. 1, which represents the solubility relations

between two components, A and B, partially miscible in the liquid state, dissolving in each other with an absorption of heat and the final disappearance of one of the liquid phases. The left-hand branch of the curve represents the solubility of A in B and the right-hand branch that of B in A. For constant temperature, below the maximum point of the curve, there are two liquid layers. At the maxi-

mum point the two layers become identical in composition. This temperature is called the critical solution temperature, and the concentration at this point is the critical concentration. The point \times represents a divariant system. At the critical concentration and constant pressure the temperature is fixed, depending only on the nature of the components. This critical solution temperature is changed by the addition of a third component C, being fixed for fixed concentration of C. This change in the critical

solution temperature by the addition of C has been applied with considerable success to the determination of the purity and composition of organic substances by Crismer,¹⁰³³ Timmermans,¹³¹⁰ Andrews,¹¹⁶⁷ and others.

Crismer¹⁰³³ found that the critical solution temperature of a binary mixture of ethyl alcohol and kerosene (considering a given

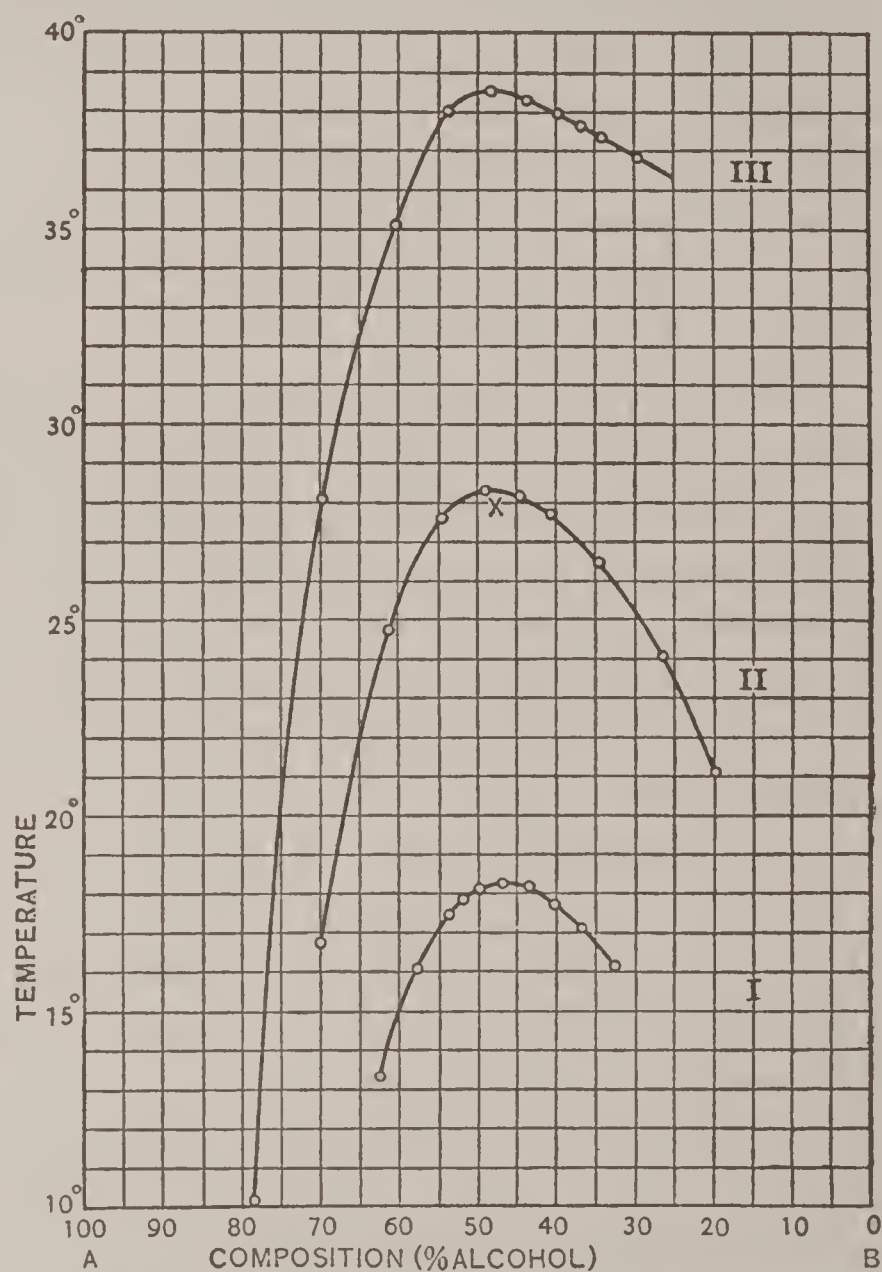


Fig. 1.—Equilibrium diagram for system ethyl alcohol-kerosene

kerosene as a component of constant composition) varied so considerably on the addition of water as a third component as to give results comparing favorably with the most careful density determinations for a criterion of the water content of an alcohol-water mixture. This work was later substantiated by Andrews.¹¹⁶⁷ In the present work the results of Crismer were again substantiated and the valuation of this constant for alcohols containing very small amounts of water proved of considerable importance in testing the fractions of several of the distillates. The variation of the critical solution temperature as a function of the amount of water present is given in curves of Fig. 2, with a comparison of our own results with those of Vandam¹¹²⁰ and Andrews¹¹⁶⁷ obtained with different kerosenes.

Curve I (Fig. 2) represents the results of Vandam and Curve IV those of Andrews. They are in very good agreement. Curves II and III show the results of the present work. Only mixtures with small amounts of water were investigated. In order to make this physical constant of value over a large range of compositions, it would be necessary to have a series of oils so chosen as to keep the critical solution temperatures within the range of ordinary temperatures most easily observed. The variation for 1 per cent of water can be read from the curves.

Curves II and III are for oils A and B, respectively (p. 347). The results indicate a large variation of the critical solution temperature caused by small quantities of water, and this variation is independent of the nature of the oil used.

As pointed out by Crismer, the temperature requires to be determined to 0°05 to give the same precision as density determinations to the fifth decimal place, which requires temperature regulation to 0°01 or 0°02 in making density determinations. As shown in Fig. 1, the critical concentration was determined by determining the solubility curves. The maxima of the solubility curves for alcohols containing small amounts of water are not shifted appreciably with respect to the alcohol-oil composition. In Fig. 1, considering the concentration of the water as being measured along an axis perpendicular to the plane of the paper, the upper Curve III represents the projection of the real

solubility curve upon the plane of the paper, which is the zero water plane.

Curve I (Fig. 1) shows the solubility curve obtained with oil B and alcohol V-2, a distillate shown in Table X. Curve II represents the solubility relations of oil A and alcohol IX-5 of Table XIII. Curve III gives the curve for oil A and an alcohol IX-0 before dehydration. Since the maximum of the curve is higher than the value given in Table XIII, the sample evidently absorbed

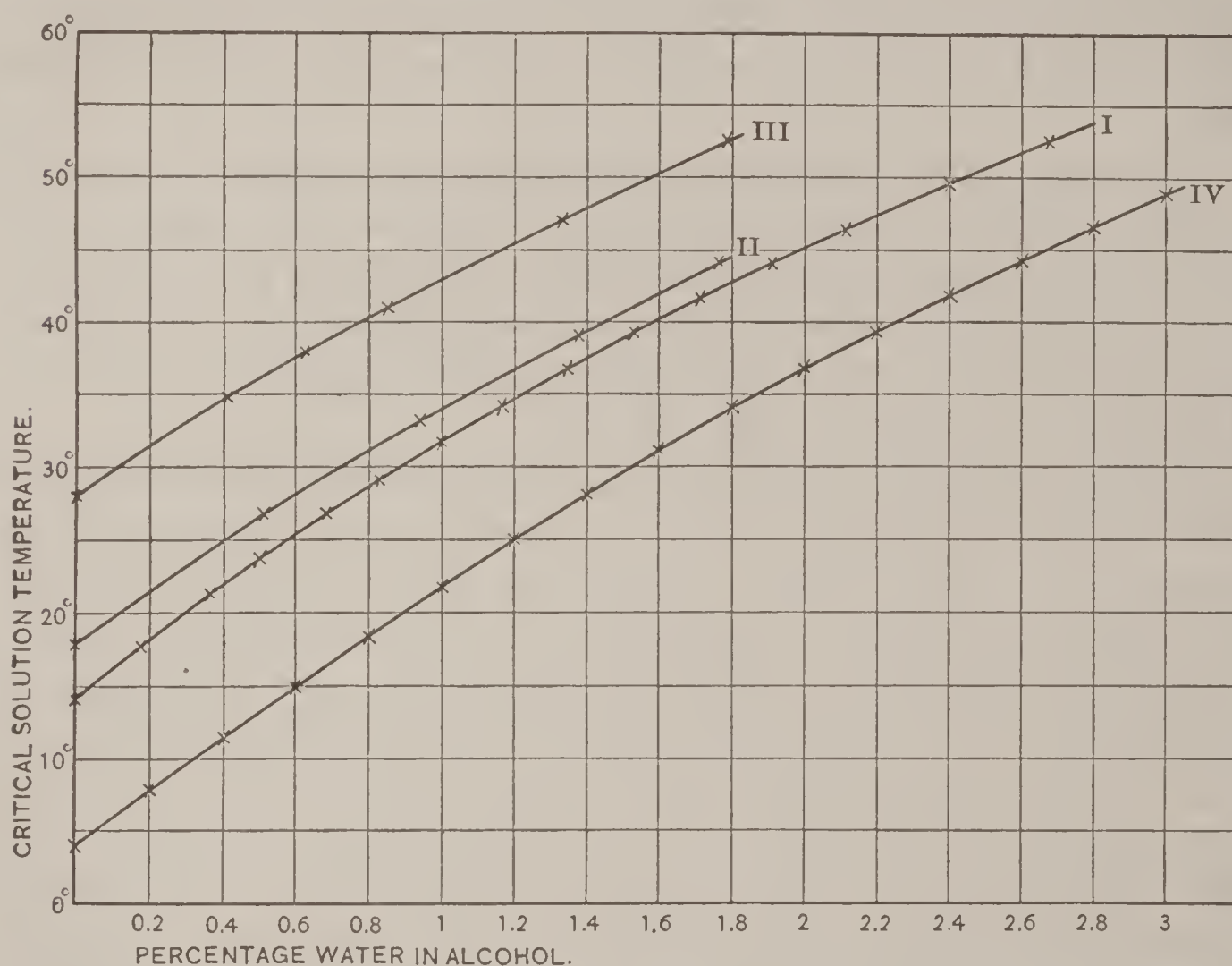


Fig. 2.—Change of critical solution temperature with varying water content of ethyl alcohol

additional moisture during the time between the two determinations.

In the determination of the critical solution temperatures equal volumes (generally 5 cc) of the alcohol and the kerosene were placed in a small bulb, care being taken to exclude moisture. This was warmed to the region of complete miscibility and slowly cooled in a bath the temperature of which could be varied at any desired rate. At the critical solution temperature there is a

sharp cloud formation. No difficulty was experienced in repeating observations to 0°05 C. In this system there was no disturbing critical opalescence at the critical concentration. In the solubility-curve determinations the end point was not so sharp for the concentrations containing a large percentage of oil.

Kerosene is especially adapted for use as one of the components, since it can be so chosen or changed by mixing with lighter hydrocarbons that the value of the critical solution temperature will be such as to permit convenient temperature measurements. Two kerosenes were used, which will be designated as oil A and oil B.

Oil A had a critical solution temperature with anhydrous alcohol of 28°3. Slow changes in this value took place when the oil was dried over sodium. It was found that a lower and more constant value could be obtained if the oil were dried over anhydrous calcium chloride. The changes are probably due to the chemical action of the sodium on the oil.

Oil B was made by diluting oil A with a lower-boiling kerosene. With anhydrous alcohol it gave a critical solution temperature of about 18°0. The oil had a density at 25° of 0.8102, was dried over anhydrous calcium chloride, and its composition as determined by check determinations with anhydrous alcohol remained unchanged over a period of several months. The preliminary experimental work showed that equal volumes of the two liquids were sufficiently near the critical concentration to permit this concentration to be used for all determinations of the critical solution temperature of alcohols containing very small amounts of water and where the relative values are of most importance. The percentage concentration by weight of alcohol in such a mixture with oil B was 49.2 per cent. Examination of the curves will show this to differ very little from the critical concentration as shown by the maxima of the solubility curves.

In certain of the experiments the values for the critical solution temperature are given in comparison with the density values.

3. MATERIALS

Purification and dehydration experiments were made on four samples of alcohol of different commercial origin. These will be

designated as samples A, B, C, and D. Sample A was purchased from Iler & Co., The Willow Springs Distillery, Omaha, Nebr., and is known to the trade as "silk-finished spirits." From density determinations, this alcohol showed a percentage strength by weight of 92.8, and it was used in most of the experimental work. An analysis made at this bureau gave the following results:

	Per cent.
Aldehydes (as acetic aldehyde).....	0.0027
Methyl alcohol (Trillat ⁸⁶³ , ⁸⁶⁴ method and Mulliken and Scudder ⁸⁹⁵ , ⁹²² , ¹⁰⁸² method).....	None.
Furfural.....	0.00015
Fusel oil (as amyl alcohol), Allen-Marquardt ³⁸⁹ method	0.033

No methyl alcohol was found by the same methods in the first distillate from 4 liters. Methods of analysis of Association of Official Agricultural Chemists ¹²⁶⁵ were used. This analysis shows the sample to have a satisfactory purity for a material with which to start a further extended purification.

Sample B, of approximately the same strength as sample A, was obtained from the Columbia Distilling Co., and showed a satisfactory purity.

Sample C consisted of Squibb's absolute alcohol, having a density of 0.78570 at 25° C., corresponding to 99.8 per cent on the basis of the Bureau of Standards tables.¹²¹⁶ This alcohol was several years old, and qualitative tests showed it to contain considerable aldehyde. It also possessed a foreign aromatic odor.

Sample D consisted of Merck's absolute alcohol Ph. IV, having a density of 0.78705 at 25° C., corresponding to 99.3 per cent. This sample contained considerable amounts of aldehyde, and was used directly in a final distillation without preliminary dehydration and purification.

4. PURIFICATION AND TESTING FOR IMPURITIES

In the preliminary purification attempts to remove the last traces of aldehyde by the method of Winkler ¹⁰⁸⁷ were unsuccessful, both at ordinary and the boiling temperature. Considerable amounts were removed in this way, but no part of the distillate was found entirely free from aldehyde. No attempt was made to remove the higher boiling impurities other than water, except by rejecting the first and last tenths of the distillates.

Sample A was treated with silver oxide in alkaline solution under the reflux condenser for several hours and then distilled. Sample B was treated in the same manner in the presence of considerable amounts of lime, in order to dehydrate at the same time. Sample C was treated with silver oxide in alkaline solution at room temperatures, the clear liquid decanted and distilled, and then subjected to the final distillation. In all preliminary dehydrations the great bulk of the impurities were rejected in the first and last tenths of the distillate. With samples A and B it was possible by means of one dehydration with lime, in the proportion recommended by Kailan ¹¹⁴⁸ (550 grams per liter), to obtain large quantities of alcohol having a density of 0.78961 at 20° C, corresponding, according to the Bureau of Standards ¹²¹⁶ tables, to 99.9 per cent alcohol by weight. This still contained minute traces of aldehyde, the impurity most easily detected. The method of aldehyde removal with metaphenylenediamine hydrochloride, suggested by Windisch ⁴⁸³, was not used because of the danger of introducing into the alcohol even more objectionable impurities. The last traces of aldehyde were successfully removed by the method of Paul ⁷⁷⁸. The alcohol was boiled for several days under a reflux condenser, coming in contact with nothing other than glass and dry air. The cooling water in the condenser was held at a temperature varying between 50° and 60° C. The top of the condenser was provided with an arrangement by means of which the more volatile constituents could be drawn off by means of a current of dry air. This arrangement is shown in Fig. 4. At the end of such a treatment, lasting two days, no more aldehyde could be found in the vapors drawn off and the most delicate tests failed to reveal it in the alcohol. During this procedure the alcohol did not change a determinable amount in density, as shown by determinations made before and after the experiment. There was some indication of aldehyde formation when the aldehyde-free alcohol was exposed to diffused daylight for a few hours. After keeping in the dark for a week, tests showed about as much aldehyde as was present before the treatment.

In general, it seemed that chemical methods for removing aldehyde depending upon its oxidation to the corresponding acid were useless in entirely removing the aldehyde. It seemed hardly

possible that an agent could be found which would oxidize aldehyde completely without attacking alcohol in the least. Agents such as silver nitrate, silver oxide, and potassium permanganate certainly fail. Metaphenylenediamine hydrochloride is very efficient in case a water-free alcohol is not desired. There has been very little work done on the efficiency of the opposite procedure—that is, reduction of the aldehyde to the corresponding alcohol. According to Wurtz¹⁸¹ ethyl alcohol can be prepared by reducing acetaldehyde with sodium amalgam. It was hoped that in the use of aluminum amalgam as a dehydrating agent it would also prove a means of removing the last traces of aldehyde. In view of the consideration that a small quantity of aldehyde is probably formed in the dehydration and in view of the above experiment and additional ones to be given later, showing its small effect on the density, it was not deemed worth while to make absolutely sure that the dehydration experiments were started with a strictly aldehyde-free alcohol.

In general, the final distillates were tested for the impurities likely to occur, such as aldehydes, the higher alcohols, ether, and water. A brief description of the procedures found most satisfactory follows.

Tests for Aldehydes.—From among the various tests for the lower aldehydes the sulphite-fuchsine reaction was found to be the most sensitive and reliable. Ammoniacal silver solution, silver nitrate solution, the Windisch reaction, using metaphenylenediamine, and the Fischer-Penzoldt⁴¹⁰ test, using diazobenzene sulphonic acid, were not found satisfactory, either because of lack of sensitiveness or of the interfering action of the alcohol itself. The sulphite-fuchsine test as proposed by Schiff²⁰⁹ and further developed by Gayon⁴⁹³ and Paul⁷⁷⁸ was found fairly satisfactory for small amounts of aldehyde in alcoholic solution. The reagent was made up according to the method of Paul⁷⁷⁸, using sulphurous acid to decolorize the fuchsine, and all tests were made with close adherence to his procedure. The relative results on the aldehyde content of the various fractions are given in some of the experiments. There is, however, some uncertainty as to the absolute amount of aldehyde present. In some fractions it was impossible to get a positive test for this impurity, and it is safe to say that

in the fractions showing the strongest test the amount of aldehyde was less than 0.001 per cent, and so of negligible effect on the density. Experiments made with water solution of formaldehyde showed that by means of this reaction one part in a million could just be detected. The sensitiveness is decreased by the presence of alcohol on account of the uncertain action of pure alcohol itself on the reagent.

Tests for Higher Alcohols (Fusel Oil).—The quantities of any higher alcohols present were so small as to escape detection by any of the ordinary quantitative methods. After the preliminary purification and one distillation no color was given on standing for a week mixed with an equal volume of concentrated sulphuric acid. It was reasonably concluded that the alcohol was free from higher alcohols in any quantity sufficient to affect the density, outside the limits of the experimental error in its determination.

Other Possible Impurities.—It was considered unlikely that ethyl ether was present, though theoretically a further treatment with a strong dehydrating agent could possibly take from the chemical compound ethyl alcohol ($\text{C}_2\text{H}_5\text{OH}$) the constituents of water, forming the ethyl oxide (C_2H_5)₂O. In the absence of any reliable chemical test for small quantities of ether in the alcohol it was concluded that any appreciable quantity would, on account of its much lower boiling point, show in the first distillate and be recognized both by odor and by a different density. It was deemed unnecessary to consider testing for such unlikely impurities as methyl alcohol and acetone which could not be found in the original samples of alcohol.

Tests for Water.—No specific chemical test for water in such small quantities as they existed in the distillates was found to be of any value. Mendeléeff^{207, 241} found barium oxide, zinc ethyl, and sodium amalgam unadapted to detect traces of moisture in alcohol. The test with lead-potassium iodide suggested by Biltz¹¹²⁹ was found to be inapplicable to alcohol. Such agents as calcium carbide, anhydrous copper sulphate, and metallic calcium gave no indication of water with alcohols whose density at 25° C was below 0.78510.

The determination of certain physical constants is the most satisfactory test for and criterion of the water content. The only

objection to such criteria is the necessary assumption of the absence of other substances in quantities sufficient to affect the physical constant determined. Making this assumption then, the determination of physical constants becomes the only satisfactory method for determining quantities of water so small. Since this work has to do with the establishment of a firm basis for density alcoholometric tables, the determination of the density was taken to be the main criterion of the water content.

A detailed description of the methods employed in the density determinations and the data on the determinations which were chosen as establishing the density of absolute alcohol will be found in part 3 of this paper (p. 405). It having been there shown that a knowledge of the condition of saturation of the alcohol with air is essential in order to use density as a criterion of dehydration, the densities given here are for alcohol saturated with air at the temperature of the determination.

Some use was also made of the determination of the critical solution temperature as a second criterion. The best criterion of the attainment of a chemically pure anhydrous ethyl alcohol was the obtaining of a considerable sample, which on further treatment with the same dehydrating agent gave a distillate with the same density from beginning to end of distillation and a value very close to that obtained from various samples of alcohol and by using various efficient methods of dehydration.

5. DEHYDRATION AND DISTILLATION OF THE ALCOHOL

Dehydrating agents.—Such reagents as potassium carbonate, sodium amalgam, anhydrous copper sulphate, and calcium chloride were rejected. Calcium carbide was not used because of the possibility of introducing organic impurities impossible to remove. On long contact with alcohol many decomposition products are formed. Lime with the addition of a small quantity of barium oxide was used in an exact duplication of Mendeléeff's ^{207, 241} treatment. The larger number of distillations were made with lime alone, using samples as obtained on the market, and also these same after being freshly ignited. Repeated distillation of the same sample of alcohol was made with this reagent. The efficiency of metallic calcium was also tested and found satisfactory

when used in small amounts. Aluminum amalgam was used for the first time in a quantitative test of its efficiency and some of the best alcohol was obtained in this way.

Apparatus and Procedure.—The various distillations were carried out in three types of apparatus. For convenience they will be designated by the letters *L*, *M*, and *N*. The first type of apparatus, *L*, was used in all preliminary distillations and some of the earlier ones of which the results are recorded. It consisted of a 5-liter Jena flask provided with a 40-cm Hempel still-head filled

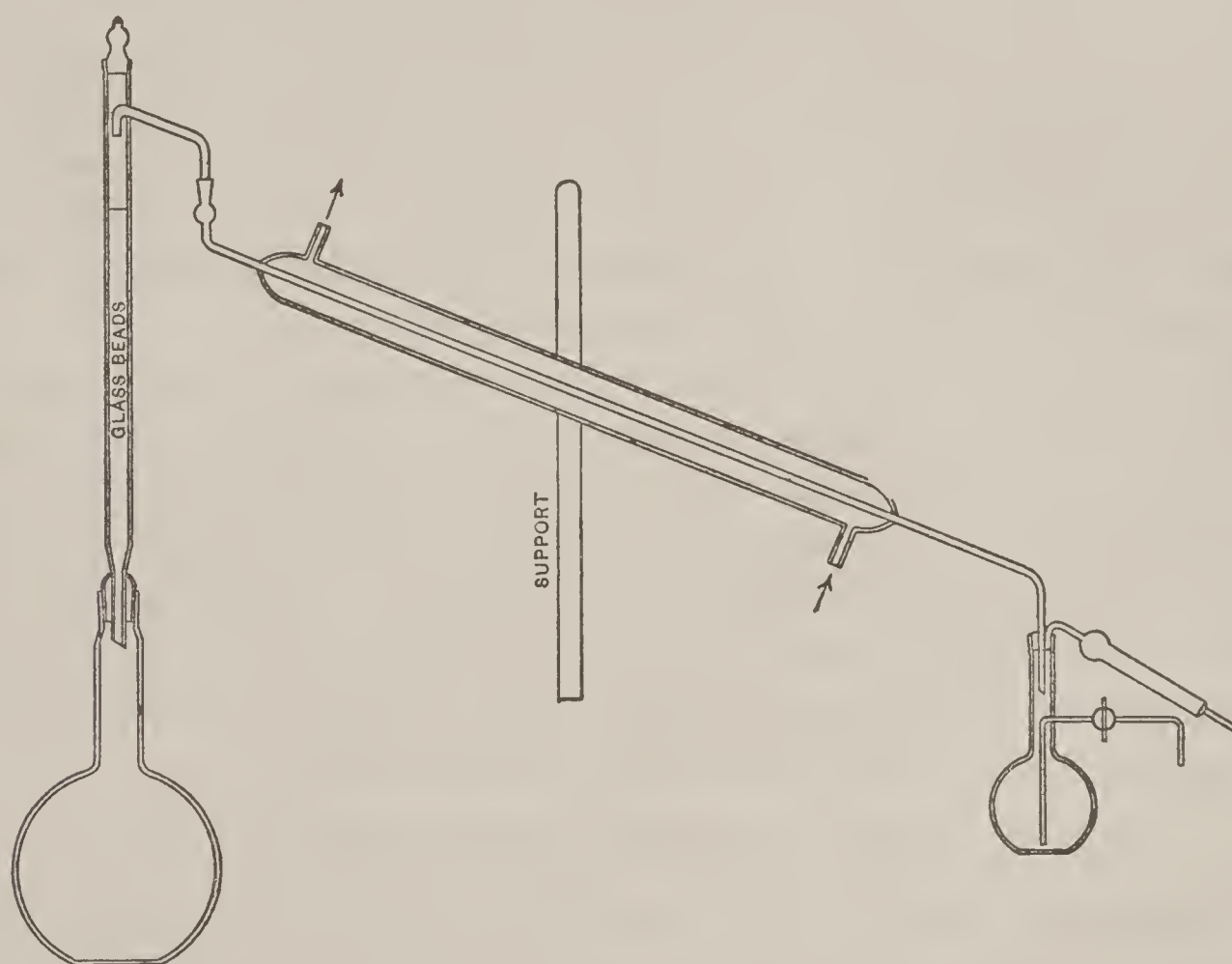


Fig. 3.—Distilling apparatus—Type *N*

with short pieces of glass tubing. To this was connected an 80-cm condenser provided with several interchangeable 500-cc flasks as receivers. These were amply protected from the moist air by means of calcium chloride tubes. The connections between the various parts were made with corks covered with tin foil and there was present the possibility of the introduction of appreciable impurities from these. The second type, *M*, was used in the distillations carried out under reduced pressure. The necessary connecting corks were in this case paraffined and the joints made

tight by means of mercury seals. The distillations were made from 2 and 3 liter round-bottomed flasks. The pressure could be reduced to 80 mm, giving a boiling temperature of about 40°C .

The third type, *N*, is shown in Fig. 3. In this apparatus the greatest efforts were made to exclude the possibility of moisture or other impurities being introduced. The still was made entirely of glass, the connecting joints being ground accurately. Distillations were made from 3- and 5-liter flasks into the 500-cc receiver. Any fraction of the total distillate under 500 cc could be siphoned off. The entire system was protected from the entrance of moisture by calcium oxide guard tubes. Fig. 4 shows the reflux condenser, used in connection with this apparatus, for the heating of the alcohol with the dehydrating agent and the carrying out of Paul's procedure of removing the aldehyde or other volatile impurities.

All preliminary distillations were made with apparatus *L*, distilling from a water bath varying in temperature between 80° and 90° . All the distillations carried out with the hope of getting an anhydrous alcohol were made from an oil bath consisting of a high grade and high boiling lubricating oil giving off no appreciable vapors below 110° . The temperature of this bath, which was heated electrically, varied in the experiments between 80° and 110° .

The following precautions to prevent moisture absorption by the alcohol were taken in all the final experiments of which the results

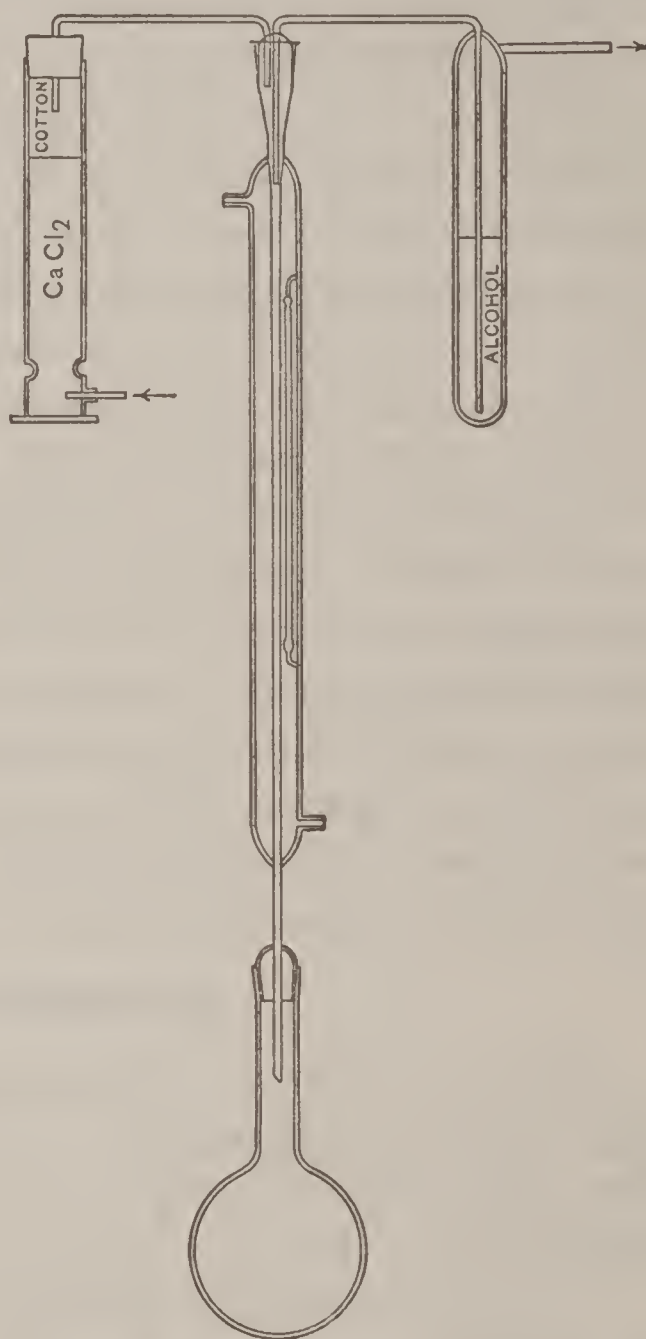


Fig. 4.—Reflux apparatus

are given. In all cases of heating under the reflux condenser the exit was provided with lime or calcium chloride guard tubes or a moving current of dry air. Immediately before putting the still head, with condenser and receiver attached, in place of the reflux condenser, the entire apparatus was swept out for 10 minutes by a stream of air thoroughly dried by passing through a calcium chloride drier, sulphuric acid, and over phosphorous pentoxide in succession. The only time during which the vapors of the alcohol came in contact with the outside air was during this transfer, which occupied about five seconds. All retainers for the distillate were swept out in the same way. The various fractions, in case they were not used at once, were always kept in tightly stoppered bottles inclosed in large lime desiccators. The alcohol at no time came in contact with moist air or any glass surface that had not been dried by means of a current of dry air. In a few distillations which will be mentioned small quantities of the dehydrating materials were placed in the still head so that the vapors of the alcohol would come in intimate contact with the same immediately before passing into the condenser. All dehydrating agents were made as nearly anhydrous as possible before any attempt was made to use them. The details of their preparation will be given under the separate experiments to be described.

6. EXPERIMENTS AND RESULTS

Fourteen separate dehydrations and distillations were made. These will be designated by Roman numerals. The following outline of the subject matter of their description is given for the sake of clearness. For each of these it is proposed to give—

1. Type of distilling apparatus used, the sample of alcohol used, its density, and, as far as needed, its previous history.
2. The preparation of the dehydrating agent, the treatment of the alcohol in dehydrating, and any special features of the distillation.
3. The results in a tabulated form, giving the number of fractions, their volume, the time occupied in their distillation, and their density at 25° compared with water at 4°, and the critical solution temperature when determined. Some density values at 15° will be given. Considering the fact that a change in density

of 0.000010 corresponds to a temperature change of only 0°012, the reader will not give undue significance to the sixth decimal place, which is here given for purposes of comparison.

4. A short discussion of the more specialized features and results of a given experiment.

Experiment I.—Three and one-half liters of alcohol from sample A were distilled in apparatus *L*. This alcohol had been dehydrated with lime and showed a density of 0.78961 at 20° C. It was also free from aldehydes. On standing for five days, protected from daylight in contact with 700 g of ordinary commercial lime, it assumed a lemon-yellow color. About 100 g of barium oxide were added and the whole heated under the reflux condenser for 10 hours. The alcohol turned a deep brown color. Immediately before the distillation 50 g of finely powdered lime were added in order to take up any water produced by the too extensive formation of barium alcoholate. This formation of water was shown by Crismer.¹⁰³³ The barium oxide was added merely with the purpose of carrying out one distillation, following as closely as possible the procedure of Mendeléeff.^{207, 241} Subsequent experiments show that it can be dispensed with, and it is certainly undesirable to have such extensive reactions, foreign to the purpose of dehydration, going on during the process.

TABLE VI
Results of Experiment I

Sample	Volume	Time	$D_{4^{\circ}}^{25^{\circ}}$
	cc	hrs	
Original.....	3500	0.78535
Fraction:			
1.....	1000	9	.785153
2.....	500	3	.785081
3.....	500	3	.785066
4.....	350	2	.785062
5.....	300	6	.785063
6.....	350	9	.785061
7.....	250	2	.785060
8.....	125	3	.785072

Though the original alcohol used was entirely free from aldehyde, small quantities were found in all the distillates with the

minimum amounts in the middle fractions. Evidently they were produced during the dehydration process. In no case was the content above 0.001 per cent. This experiment is a very satisfactory reproduction of the work of Mendeléeff, the results agreeing with his as to the value of the density, but at the same time showing a much greater uniformity in the various fractions. While the last fraction was being distilled the oil bath was at a temperature of 100°. This fact, in conjunction with later experiments, shows that the temperature of the heating bath has little to do with the composition of the distillates when distilling a nearly anhydrous alcohol, provided it does not exceed 110°. Within the limits tried the rate of distillation seems to have no disturbing influence. The rate was varied from 40 to 175 cc per hour without any significant change in the density values.

Experiment II.—Three liters of sample A, having a density of 0.78961 at 20° C, after preliminary dehydration, were treated with lime, duplicating as closely as possible the procedure of Messrs. Squibb.⁴³³ The distillation was carried out in apparatus *M* under reduced pressure, varying between 80 and 100 mm, necessitating a bath temperature between 35° and 45°. Air-slaked lime in powdered condition was ignited at a red heat for 10 hours and let cool in dry air. This, with some large pieces of freshly ignited lime, aggregating about 400 g per liter of alcohol, was rotated in the cold for three days and let stand for six weeks with frequent shaking. The clear lemon-colored liquid was siphoned off and distilled.

TABLE VII *
Results of Experiment II

Sample	Volume	Time	$D_{4^{\circ}}^{25^{\circ}}$
	cc	hrs	
Original.....	3000	0.78535
Fraction:			
1.....	500	1	.785097
2.....	500	1	.785088
3.....	500	3	.785085
4.....	500	3	.785107
5.....	500	3	.785101

The fractions from this distillation contained traces of aldehyde. By dehydration and treatment essentially similar to that described

by Messrs. Squibb, it was not possible to duplicate the low values gotten by them for the density of anhydrous alcohol. The values, on the other hand, agree with those obtained by other investigators using a great variety of dehydrating agents. The slightly higher values of all the fractions may be due to their containing less dissolved air.

Experiment III.—One liter of lot C of Squibb’s manufacture, after the preliminary purification had reduced the foreign odor and removed the bulk of the aldehyde content, was treated with 400 g of freshly ignited lime in apparatus N. It was subjected to Paul’s ⁷⁷⁸ treatment in order to remove any of the more volatile constituents still remaining.

TABLE VIII
Results of Experiment III

Sample	Volume	Time	$D_{\frac{25}{4}}^{\circ}$
	cc	hrs	
Original.....	1000	0. 78570
Fraction:			
1.....	500	3	. 785072
2.....	200	1	. 785070

The results of this experiment show the efficacy of Paul’s procedure in removing the lower boiling impurities. In the distillates the foreign odor and the aldehyde were just noticeable. The values are very close to what appears to be the normal one.

Experiment IV.—This experiment was intended to represent our best efforts in obtaining a pure anhydrous alcohol and was carried out with every precaution that occurred to us. Three and three-tenths liters of mixed distillates from previous experiments were treated in apparatus N with 500 g of lime made from marble. This lime was ignited for 10 hours in an electrical resistance furnace at from 600° to 700°. After the mixture had been left standing for three days at room temperature, with frequent shaking and after the addition of 25 g of powdered lime that had been ignited to 900° for 24 hours, the whole was heated under the reflux for 10 hours. The same amount was added immediately before distilling. A considerable quantity of small

chips, ignited in the same way, was put into the still head and the distillation begun before the alcohol had cooled. Fractions were taken directly from the receiver into the tube in which the density determinations were made. The distillate was so divided into fractions as to show the possible differences in the physical properties of its parts to best advantage.

The results show the distillate to have been very nearly uniform throughout. The density at 25° of fraction 2 on keeping for several months in a glass-stoppered bottle in a lime desiccator that was frequently opened changed to 0.785088, representing an absorption of 0.006 per cent moisture, assuming no other changes in the alcohol of such a nature as to influence the density.

TABLE IX
Results of Experiment IV

Sample	Volume	Time	$D_{4^{\circ}}^{25^{\circ}}$
	cc	hrs	
Original.....	3300	0.785116
Fraction:			
1.....	120	0.5	.785097
A.....	450	1.5	
2.....	120	.5	.785069
B.....	500	1.0	
3.....	120	.5	.785058
C.....	350	.5	
4.....	120	.5	.785056
D.....	350	1.0	
5.....	120	.5	.785050
E.....	400	1.0	
6.....	450	1.0	.785054

Omitting the density value of the first fraction the various fractions have the same density to within two units in the fifth decimal place.

Experiment V.—Four liters of sample B, comprising the middle fractions from the preliminary dehydration and purification, were added to the residues and lime of Experiment IV in the same apparatus.

Heating under the reflux was continued for a week with the water in the reflux between 50° and 60°. The still head containing the chipped lime was again used.

TABLE X
Results of Experiment V

Sample	Volume	Time	$D_{4^{\circ}}^{25^{\circ}}$
Fraction:	cc	hrs	
1.....	400	1	
2.....	1000	3	0. 785051
3.....	2100	4	. 785101
4.....	200	1	

These distillates contained no aldehyde. They were used in subsequent experiments to test the effect of successive dehydrations and the use of calcium. Freshly ignited lime does not seem to be necessary in order to obtain an alcohol of very low density.

Experiment VI.—The third fraction of 2000 cc from Experiment V was distilled from apparatus *N*. This alcohol was treated with about 400 g of commercial lime taken from large pieces and the whole heated under the reflux for 18 hours. The middle fraction of 1250 cc showed a density of 0.785058 at 25°.

It appears from this experiment that a repetition of the dehydration even with commercial lime reduced the density of the alcohol product considerably.

Experiment VII.—The middle fraction of Experiment VI was treated with 150 g of chipped lime that had been ignited at 900° for 10 hours. This was heated under the reflux for 18 hours and then distilled in apparatus *N*.

TABLE XI
Results of Experiment VII

Sample	Volume	$D_{4^{\circ}}^{25^{\circ}}$
Original.....	cc 1250	0. 785058
Fraction:		
1.....	120	. 785064
A.....	250	
2.....	120	. 785059
B.....	120	
3.....	120	. 785060

This is the final of three successive dehydrations of the same sample of alcohol with lime. The value remains essentially unchanged and the separate fractions show a remarkable agreement in their densities, indicating the purity and uniformity of the alcohol. It seems that this distillate represents the best results obtainable with lime as a dehydrating agent, and also from the standpoint of uniformity of the different parts of the distillate. It is probable that this alcohol is freer from impurities than any obtained in this work with the possible exception of the distillates of Experiment XII.

Experiment VIII.—The combined fractions from Experiment VII, aggregating about 750 cc, were treated in apparatus N, with 6 g of metallic calcium and let stand over night. The calcium acted upon the alcohol with hydrogen evolution giving a white crystalline alcoholate, which went into solution on raising the temperature.

TABLE XII
Results of Experiment VIII

Sample	Volume	Time	$D_{4^{\circ}}^{25^{\circ}}$
	cc	hrs	
Original.....	750	0.785061
Fraction:			
1.....	250	3	.785081
2.....	250	1.5	.785072
3.....	200	2	.785080

The table shows a slight increase in the density results. This can not be certainly ascribed to an increase in the water content. It may easily be due to a variation in the condition with regard to air saturation caused by the calcium treatment or to the formation of a small quantity of some other substance. The residues in the flask were brownish. The distillation was carried almost to dryness. Other investigators have found smaller amounts of calcium more efficient than large amounts. The value obtained for the density by use of this agent is lower than that obtained by other investigators, excepting Klason and Norlin.¹¹⁰⁹ Results indicate that lime is more efficient than metallic calcium.

Experiment IX.—Four liters of sample D without preliminary dehydration and purification were treated with 400 g of recently ignited lime per liter of alcohol. This was let stand for several weeks with frequent shaking. As in the other cases of dehydrating with lime, a lemon yellow color appeared, deepening finally to a brown. No attempt was made to remove the aldehyde by chemical means. The whole was subjected to Paul's ⁷⁷⁸ treatment of boiling under the reflux for about two days. Distillation was carried out in apparatus *N* after adding about 25 g of freshly ignited powdered lime.

TABLE XIII
Results of Experiment IX

Sample	Volume	Time	$D_{4^{\circ}}^{25^{\circ}}$	Critical solution temperature
	cc	hrs		°C
Original.....	4000	0. 787048	37. 60
Fraction:				
1.....	120	1. 5	. 785128	18. 45
A.....	500	1. 5
2.....	150	. 5	. 785078	18. 25
B.....	500	1. 5
3.....	200	. 5	. 785067	18. 30
C.....	500	2. 0	18. 25
4.....	120	. 5	18. 10
D.....	500	3. 0	18. 20
5.....	150	3. 0	. 785048	18. 17
E.....	500	3. 5	18. 15
6.....	500	6. 5	. 785101	18. 30

In the last column are given the values of the critical solution temperature. The relative values of this constant found for the different fractions are somewhat irregular, but in general follow the density values. By one dehydration with lime of a commercial absolute alcohol as above it is possible to get a middle portion of the distillate showing the density value of anhydrous alcohol. This alcohol from a different commercial source furnishes anhydrous alcohol of essentially the same physical properties as the other samples examined.

Experiment X.—The three following experiments were made, using activated aluminum (aluminum amalgam) as dehydrating agent. There are several statements in the literature regarding its efficacy, but no quantitative experimental data.

One liter of sample A, having a density of 0.78535 at 25°, after the preliminary dehydration, was treated with 10 g of aluminum amalgam. The distillation was carried out in apparatus *M* under reduced pressure. The amalgam was prepared as recommended by Wislicenus and Kaufmann⁷⁵⁰ by etching aluminum wire with sodium hydroxide solution and then treating with a dilute solution of mercuric chloride. The amalgam, after washing with alcohol and ether and subsequent drying, was let stand with the alcohol over night. There was an abundant formation of the insoluble alcoholate with copious hydrogen evolution.

TABLE XIV
Results of Experiment X

Sample	Volume	Time	D ^{25°} _{4°}
	cc	hrs	
Original.....	1000	0.78535
Fraction:			
1.....	500	4	.785112
2.....	300	2	.785077
3.....	100	1	.785088

Twenty cubic centimeters of fraction 2 were added to fraction 3 to give sufficient volume for the determination of the density.

Experiment XI.—One liter of the same sample as used in the previous experiment was treated with 10 g of amalgam and let stand for three weeks. This amalgam was made by heating the wire in mercury up to the boiling point of the latter. The distillation was carried out in apparatus *M* under reduced pressure. After rejecting the first 50 cc the middle fraction of 400 cc showed a density, at 25°, of 0.785089.

Experiment XII.—Two and one-half liters of sample A, after preliminary purification, were treated with amalgam made from 25 g of aluminium wire. The alcohol was kept boiling for a week under a reflux condenser containing water varying in temperature between 50° and 60°. No aldehyde was contained in the vapors drawn off at the end of this treatment. Immediately before distilling in apparatus *N* some freshly amalgamated wire was placed in the still head.

TABLE XV
Results of Experiment XII

Sample	Volume	$D_{4^{\circ}}^{25^{\circ}}$
Original.....	cc 2500	0.78535
Fraction:		
1.....	250	.785056
2.....	250	.785068
3.....	500	.785051
4.....	350	.785051
5.....	250	.785061
6.....	300	.785056

The alcohol obtained in this experiment represents a pure liquid within the limits set by the sensitiveness of the density determination as a criterion of purity. The density results agree very closely with those obtained by other methods of dehydration. All the fractions were practically free from aldehyde. The amalgam may act as a reducing agent, converting aldehyde into the corresponding alcohol or at least preventing any formation of aldehyde during the dehydration.

Experiment XIII.—In order to furnish anhydrous alcohol for a check series of density determinations of mixtures given in part 4, two additional distillations were made.

Two liters of a mixture made from fractions of various distillations of lots A and B (IV, 1; V, 4; VI, 1; XII, 1, 6) were treated with 200 g of freshly ignited lime. The alcohol was let stand for 10 days with frequent shaking. The clear liquid was siphoned off with careful exclusion of moist air and treated under the reflux condenser for a day after adding 50 g of freshly ignited, air-slaked lime which was in a very fine state of division. The water in the reflux was held at about 60° and the more volatile constituents were removed by means of a moving current of dry air led into the top of the condenser. Fifty grams additional of powdered lime were added immediately before distilling. Apparatus *N* was used with the receiver directly sealed to the condenser. The densities of the various fractions were determined at 15° in order to give a direct comparison with the results of Mendeléeff. The critical solution temperature was followed closely.

TABLE XVI

Results of Experiment XIII

	Volume	$D_{4^{\circ}}^{15^{\circ}}$	Critical solution temperature
	cc		°C
Original.....	2050	17.95
Fraction:			
1.....	20	17.95
2.....	120	0. 793619	17.83
3.....	250	17.82
4.....	120	. 793608	17.82
5.....	130	17.82
6.....	150	. 793614	17.85
7.....	250	17.85
8.....	250	. 793615	17.78
9.....	50	17.75

Fraction 4 near the middle of the distillate shows the lowest density. The critical solution temperature showed a small decrease as the distillation proceeded and indicated a decreasing water content on the assumption of the absence of other impurities. The differences are hardly significant and the results show a satisfactory agreement with the other experiments. The density of absolute alcohol at 15° is somewhat higher than the corrected result of Mendeléeff.

Experiment XIV.—Fractions from the various distillations (V, 2; IX, 2, 6, C; XIII, 2, 4, 8, 9) were combined and treated with lime as given under Experiment XIII. The same distilling apparatus was also used. Density determinations were made at 15° and 25°.

TABLE XVII

Results of Experiment XIV

Sample	$D_{4^{\circ}}^{15^{\circ}}$	$D_{4^{\circ}}^{25^{\circ}}$	Critical solution temperature
			°C
Fraction:			
1.....	17.10
2.....	17.10
3.....	0. 793596	0. 785049	17.10
4.....	. 793595	. 785058	17.05
5.....	17.05
6.....	

Judging from the results obtained in the determination of the critical solution temperature, there is practically no separation of the alcohol into fractions of different physical properties. The densities obtained are slightly below the mean values obtained in the previous experiments. It is interesting to compare the expansion of fractions 3 and 4 between 15° and 25° with the value called for by the formula obtained in part 2. Here the value for $D_{\frac{15^{\circ}}{4}}^{\circ}-D_{\frac{25^{\circ}}{4}}^{\circ}$ is found to be 0.008547 for fraction 3 and 0.008537 for fraction 4, while the formula difference is 0.00854.

7. EFFECT OF IMPURITIES ON THE DENSITY

A study was then made of the effect of small concentrations of possible impurities on the density of anhydrous alcohol and on the critical solution temperature with kerosene mixtures. The effect of acetaldehyde and ethyl ether was studied. Some data were also obtained on the effect of dissolved air. (See p. 407.)

The lower aldehydes, principally acetaldehyde, are the most persistent impurities in alcohol, notwithstanding their much lower boiling points. While tests showed that acetaldehyde was not present in amounts sufficient to affect the density appreciably, yet it was deemed worth while to determine the direction and amount of this effect. Acetaldehyde was made by oxidizing alcohol with chromic-sulphuric acid mixture, then purified and dehydrated by distilling twice from anhydrous calcium chloride. Immediately before making the mixtures the vapor was passed over calcium chloride before condensation. The mixtures were made up by weighing directly. Table XVIII shows the results obtained.

TABLE XVIII
Effect of Acetaldehyde on Density

Mixture	Acetaldehyde	$D_{\frac{25^{\circ}}{4}}^{\circ}$	Increase in density
Anhydrous alcohol XII ₃	Per cent 0. 00	0. 785101	0. 000000
1	. 023	. 785144	. 000043
2	. 140	. 785394	. 000293
3	. 466	. 786030	. 000929

The aldehyde in mixture 2 could be plainly recognized by odor. Mixture 1 diluted with alcohol to 10 times its volume still gave a strong test for aldehyde by means of the sulphite-fuchsine reaction. Ramsay ³³⁸ found 0.7771 to be the density of acetaldehyde at 21°, this temperature being its boiling point. Small variations of the density of the fractions in certain experiments may be due to small quantities of aldehyde. It may be interesting to note the concordance in the various fractions of Experiment XII where aluminum amalgam was used as dehydrating agent and might act as a reducing agent converting the aldehyde into alcohol. One per cent aldehyde lowers the critical solution temperature 0°.5.

Very small quantities of ether are not open to detection by chemical means because of its resistivity toward chemical reagents in general. One would expect the bulk of any ether present to collect in the first fraction on distillation and to be recognizable either by odor or its effect on the density or other physical properties. A few mixtures of known ether content were made up and their density determined. The ether was purified by shaking with dilute sodium hydroxide solution and then washed thoroughly with water to remove the alcohol. After several days' drying over anhydrous calcium chloride it was distilled from sodium wire immediately before use. The mixtures were made up by direct weighing.

TABLE XIX
Effect of Ethyl Ether on Density

Mixture	Ether	$D_{4^{\circ}}^{25^{\circ}}$	Change in density
Anhydrous alcohol, IV ₅	Per cent		
1	0.00	0.785091
2	.060	.785070	—0.000021
	.185	.785019	— .000072

For small amounts of ether in alcohol the results show that 0.1 per cent lowers the density 0.00004. Both mixtures on standing gave a strong odor of ether. Young ⁹⁸⁶ suggests the possibility that the low density results obtained by Messrs. Squibb ⁴³³ may be due to the presence of ether. Assuming the same rate of

decrease to hold and taking 0.78506 as the correct value for the density of anhydrous alcohol at 25°, it would take approximately 0.9 per cent ethyl ether to give their low value. This amount could hardly escape detection. One per cent ether lowers the critical solution temperature 2°1.

8. DISCUSSION OF RESULTS

The distillates obtained in Experiments IV and XII answer all the requirements as to purity, judged by the constancy of the physical property determined. That they were not mixtures showing constant physical properties is indicated from the fact that the density results agreed even when different dehydrating agents were used. That the density of the distillate remained essentially unaltered when the distillation was carried on under reduced pressure is additional evidence on this point.

Concerning the freedom from impurities other than water in the distillates showing densities, at 25°, of 0.78506 ± 0.00001 , the following is to be said: (1) The fact that fractions of the same density were obtained from samples of different origin points to either a removal of all the significant amounts of impurities or a retention of these impurities in fixed amounts even though using different methods of purification; (2) the good agreement found in the density of the mixtures made up from the distillates and mixtures of distillates with densities showing water content from 0.01 to 0.05 per cent, and from different alcohols, indicates a uniformity in the alcohol characteristic of a pure substance within our present methods of determination.

III. SUMMARY OF RESULTS

1. No difficulty was experienced in obtaining alcohol containing only one-tenth of 1 per cent of water by dehydrating with lime. The dehydration proceeded more rapidly at the boiling temperature of the alcohol. The bulk of the impurities other than water were removed by the rejection of the first and last tenths of the distillate in the preliminary distillation.

2. Chemical methods for removing the last traces of the lower aldehydes were found to be unsatisfactory. This was accomplished by the method of Paul.⁷⁷⁸ Results indicated that minute quan-

tities of aldehydes are produced during the procedure of dehydration and distillation. These quantities can just be detected chemically and were so small as to have no measurable effect on the physical constants determined.

3. An alcohol was obtained which on a further dehydration was not lowered in density and on distillation gave fractions showing the same density within the experimental errors of the determination. Practically the same density value, 0.78506 at 25°, was obtained by using calcium oxide and activated aluminum, and this value is only slightly lower than the results of the more careful previous experimenters. The two dehydrating agents mentioned were equally satisfactory in removing the last traces of water. Slightly higher density values were obtained when calcium was used.

4. By a repetition of Mendeléeff's ^{207, 241} procedure used in dehydrating alcohol a distillate showing about the same density value was obtained. The various fractions of the distillate showed greater uniformity in density than in his work. A repetition of the work of Messrs. Squibb in its essential details failed to produce alcohol showing the same low density values obtained by them.

5. Small amounts of acetaldehyde increased, while small amounts of ether decreased the density of ethyl alcohol. It was shown that Squibb's ⁴³³ low density values can not be ascribed to the presence of ethyl ether.

6. In substantiation of the work of Crismer ¹⁰³³ and Andrews ¹¹⁶⁷ the value of the critical solution temperature as a method for detecting small amounts of water in alcohol was demonstrated and shown to have approximately the same sensitiveness as density determinations to one unit in the fifth decimal place.

PART 2

THERMAL EXPANSION OF MIXTURES OF ETHYL ALCOHOL AND WATER

By N. S. Osborne

Part 2 consists of the determination of the thermal expansion between 10°C and 40°C of 12 mixtures of alcohol and water, and the derivation of the coefficients for calculating, between these limits of temperature, the density of any mixture of alcohol and water.

IV. DESCRIPTION OF EXPERIMENTAL WORK

1. METHOD OF DETERMINATION AND GENERAL DESCRIPTION OF APPARATUS

Twelve mixtures whose thermal expansions were investigated were made of approximately integral percentages for convenience, and the concentrations chosen were such as would facilitate interpolation of results. The density of each mixture was twice determined at each of the following temperatures: 10° , 15° , 20° , 25° , 30° , 35° , and 40°C , one series starting with the lower and one with the higher temperature. The density determinations were made by the hydrostatic weighing method—that is, by weighing in the liquid a sinker of known mass and volume.

The arrangement of apparatus is shown in Figs. 5, 6, 7, and 8. The densimeter tube containing the liquid under investigation and the immersed sinker *E* is shown in Fig. 5. A special cap was used for closing the densimeter tube when weighings were not being made. This cap consists of a brass cover “a,” fitted to the tube by a soft rubber bushing. Through the center of the cover is a hole, which may be closed by a tightly fitting brass plug. To the upper and lower faces of this plug is attached the suspension wire. The arrangement is shown in Fig. 5. When weighings are in progress, the suspension is as shown at the right, access of the outer air being only through the hole in the cap. When not

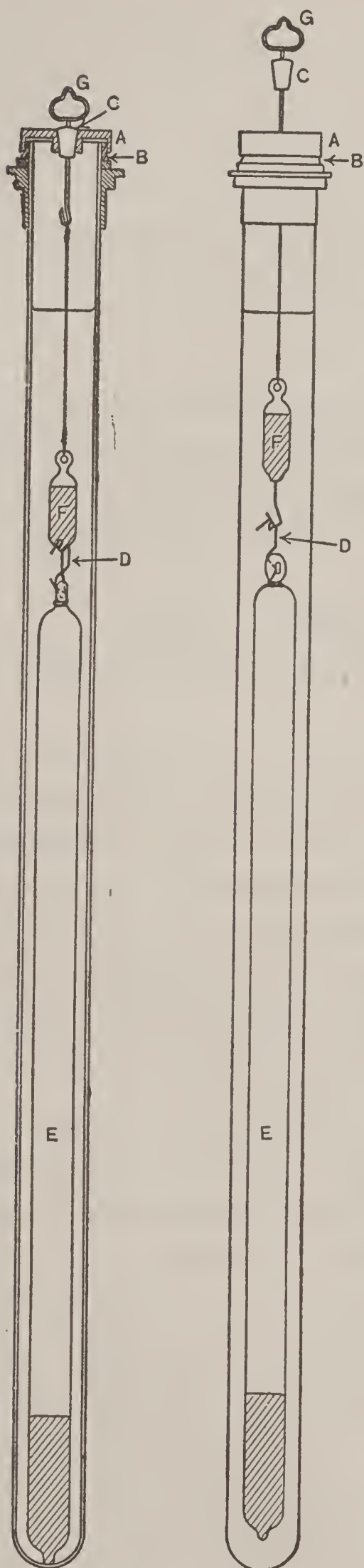


Fig. 5.—Densimeter tube
(one-third size)

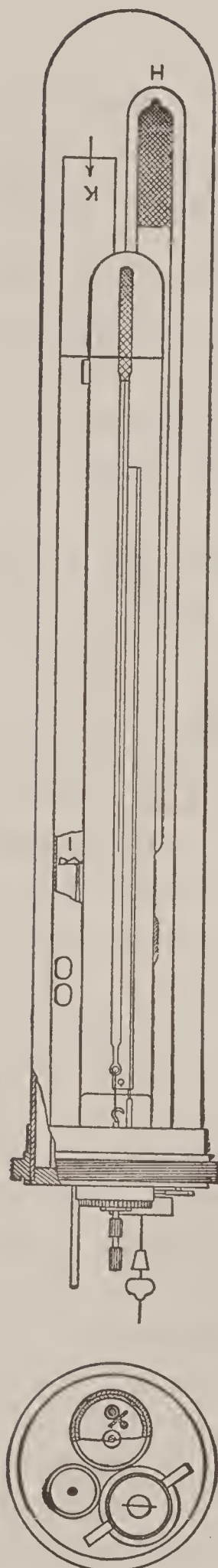


Fig. 6 (one-fourth size)

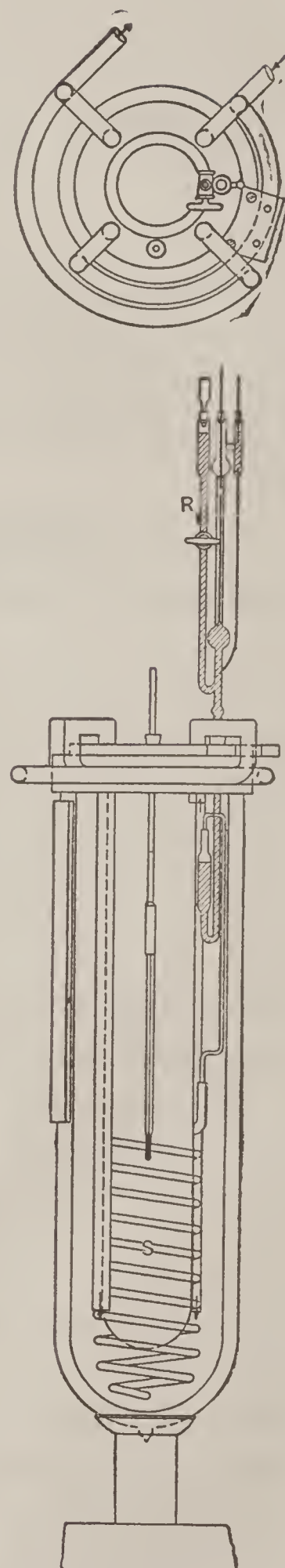


Fig. 8 (one-eighth size)

observing, this hole is closed by the plug, as shown at the left, in order to avoid unnecessary changes in concentration of the liquid, caused by evaporation of the alcohol or by absorption of water vapor from the air. In Figs. 6 and 7 the densimeter tube *H* is shown fixed in place in the inner water bath.

The water in the inner bath is kept in constant circulation by the propeller *I*. This bath is immersed in an outer bath kept in constant circulation by means of a motor-driven turbine *N* placed outside the bath. The temperature of this outer bath is maintained constant or changed at will by means of the electric heating coil *O* surrounding the return pipe and by the tubular coil *P* connected with the refrigerating brine supply maintained at a temperature below 0° C. The flow of brine in this coil is adjusted by means of a valve, not shown. The flow of water in the circulating apparatus may, by means of the special valve *Q*, be directed entirely through the cooling chamber, directly through the circulating turbine, or divided, part going either way at will, thus regulating the quantity of heat removed from the system by the brine. The cooling is made slightly in excess of the heat acquired from the surrounding air and that produced by the circulation, the balance being maintained by means of the heating coil.

This coil, which has a resistance of 10 ohms, is made of "advance" ribbon, wound over mica on the brass tube through which the return flow takes place. It is used both for regulation of temperature and for rapid heating when changing the temperature. For regulation it is connected in series with a variable resistance composed of a sliding contact wire rheostat and a bank of parallel connected lamps. As a shunt on this variable resistance is connected another lamp bank in the circuit of which is a relay operated by the thermo regulator *R*. The bulb of this thermo regulator is the tubular copper coil *S*, containing about 40 cc of xylol.

A rise in temperature closes an electric circuit, the relay is thus energized, the shunt circuit broken, and the current in the heating coil diminished. When the temperature falls sufficiently, this action is reversed. The sensibility of the regulation varies according to the conditions. Under the most adverse conditions the regulating energy used in the heating coil was from 100 watts

to 140 watts and maintained the outer bath constant within $0^{\circ}.05$ C. Under the best conditions the energy varied from 10 to 20 watts, keeping the temperature of the bath constant within $0^{\circ}.01$ C. The periodic variation in temperature of the outer bath was in either case far too rapid to produce any perceptible change in the inner bath. For rapid heating when changing the temperature in the ascending series, the small regulating current was replaced, using a double-throw switch, by a heavy current supplying about 1500 watts in the heating coil.

The outer bath as first arranged is shown in Fig. 7. It is contained in a rectangular tank with plate-glass walls and brass bottom and top. The insulation consisted of two layers of heavy cardboard with cotton between. The water enters at *T* and leaves at *U*, thus insuring complete circulation. Owing to the repeated difficulty on account of leakage of the joints, this tank was replaced by a double-walled glass cylindrical vacuum jacket as shown in Fig. 8. The top was closed by a brass cap cemented to the glass. The water entered by two tubes extending to the bottom and left by two tubes at the top. The greatest difficulty, with this container for the outer bath was the distortion of the image of the thermometers, owing to the cylindrical surface. This was remedied by cementing a plane glass plate to the outside surface, forming a cell which when filled with water corrected most of the aberration. To exclude radiation the cylinder was covered with nickel paper. This vacuum jacket was used principally for the determinations of density of the mixtures of known proportions at constant temperature and was employed for the thermal expansion of only one mixture, 5 III.

The temperatures were observed on two mercury thermometers suspended in water in a tube placed in a position in the inner bath symmetrical to that of the densimeter tube. On Fig. 8 only the upper part of this tube is shown. The holders for the thermometers permitted their rotation and also allowed either of the two to be brought into position for reading. They were read by means of a microscope of long focus which could be moved vertically by means of a rack and pinion.

The reason for placing the thermometers in a tube of water instead of directly in contact with the inner bath was to minimize

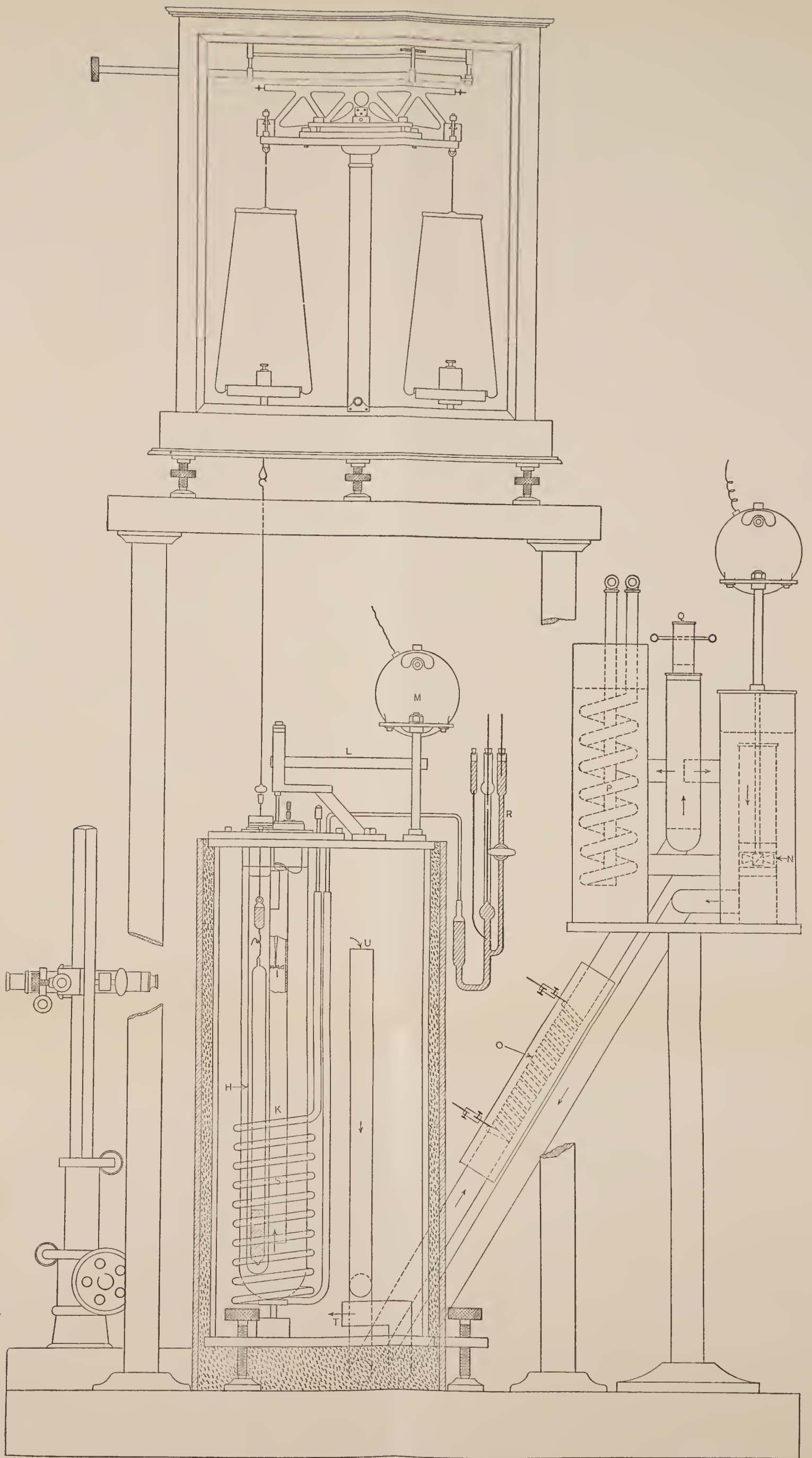


Fig. 7.—Apparatus for determination of density and thermal expansion (one-sixth size)

the error from temperature lag when bringing the temperature to constancy before observing. The containing tube was slightly larger and thicker walled than the densimeter tube; hence when the thermometers indicated a constant temperature it could be assumed that the liquid in the densimeter tube was at the same constant temperature. In the observations for thermal expansion economy of time did not permit an absolutely constant temperature to be attained, but the observed variation during a density observation extending over about six minutes never exceeded 0°.02 and, with but few exceptions, was always rising. Thus the relative error due to lag in the temperatures of the determinations was small, probably less than 0°.01 except in few instances, and may be regarded as accidental and eliminated in the adjusted results. Examinations of the residuals obtained justifies this assumption.

2. THERMOMETERS USED

Five thermometers were used in this work; they are described in Table XX.

TABLE XX

Thermometer No.	Maker	Date of manufacture	Kind of glass	Range of scale	Length of 1°	Construction
4653	Tonnelot....	1888	Verre durre..	-2°.9 C to +40°.5 C.....	mm 6.3	Solid, clear stem
2040	Haak.....	1906	Jena 16 ^{III}	{ -1 to +1° C..... +10 to +31° C..... }	4.8	{ Inclosed, white scale
264	Richter.....	1902do.....	{ -0.4 to +0°.4 C..... +31.5 to +44° C..... }	8.0	Do.
2499	Green.....	1907do.....	{ -0.5 to +0°.5 C..... +9 to +31°.5 C..... }	7.5	{ Solid, white back stem
15938	Baudin.....	1903	Verre durre..	{ +31.5 to +32°.5 F..... +89.1 to +115°.6 F..... }	7.2	Do.
15940do.....	1903do.....	{ +31.5 to +32°.5 F..... +88.9 to +115°.8 F..... }	7.2	Do.

All the thermometers were subdivided in tenth-degree divisions. No. 4653 was repaired in 1909 and the bulb reannealed. This thermometer was used in all of the experimental work at every temperature. Nos. 15938 and 15940 were used only for the thermal expansion of 5 III at 35° and 40° C. For this same mixture No. 2499 was used at 10°, 15°, 20°, 25°, and 30° C. For the ther-

mal expansion of all the other mixtures No. 2040 was used at 10°, 15°, 20°, 25°, and 30° C, and No. 264 was used at 35° and 40° C. Every temperature was observed on two thermometers, one always being No. 4653.

The thermometers were calibrated at the points 10°, 15°, 20°, 25°, 30°, 35°, and 40° C by comparison with the primary mercurial standards of the bureau. In the use of the thermometers their ice points were observed after the determination at each temperature, and the corrections to be applied to the observed temperature were deduced from the ice point reading and the calibration corrections. The corrected temperatures are in accordance with the international hydrogen scale.

3. DESCRIPTION OF SINKER

The sinker used in all hydrostatic weighings in this article is designated No. 7. It is made of Jena 16^{III} glass. The length over all is 33 cm; the outside diameter is 13 mm. The platinum hook *D*, weighing 0.5327 g, while not permanently sealed to the ring, is considered as a part of the sinker as regards mass and volume. This sinker, which is ballasted with mercury, was made in May, 1908, and was annealed at 450° C before sealing.

The mass as determined at intervals during the progress of the work is given in Table XXI.

TABLE XXI

Determinations of Mass of Sinker No. 7

	Observed mass (grams)
October 22, 1909.....	99. 9991
Do.....	99. 9991
Do.....	99. 9990
Do.....	99. 9990
March 19, 1910.....	99. 9994
Do.....	99. 9987
Do.....	99. 9990
Do.....	99. 9989
Do.....	99. 9988
Do.....	99. 9989
June 16, 1910.....	99. 9991
Do.....	99. 9986
Do.....	99. 9991
Do.....	99. 9989
Mean.....	99. 9990

The volume of the sinker was determined by weighing in distilled water from which the air had been removed. Table XXII gives the determinations of volume made previous to the use of sinker. The results are based on the value of Chappuis ¹¹³² for the expansion of water, using 99.9990 g for the mass of the sinker.

TABLE XXII

Determinations of Volume and Thermal Expansion of Sinker No. 7

Date	Observed mean tem- perature	Observed mean volume	Reduction to integral temperature		Calculated V _t	Observa- tions
			t	V _t		
1909		ml		ml	ml	
Oct. 22.....	3. 93	47. 6958	4	47. 6957	47. 6956	8
Oct. 21.....	10. 247	47. 7024	10	47. 7019	47. 7020	8
Do.....	20. 136	47. 7131	20	47. 7128	47. 7128	8
Do.....	30. 080	47. 7242	30	47. 7239	47. 7238	8
Do.....	39. 884	47. 7349	40	47. 7348	47. 7350	8
Oct. 22.....	40. 022	47. 7355	40	47. 7353	47. 7350	8
			25	47. 7183	Calcu- lated

Equation for calculation of volume at any temperature:

$$V_t = 47.7172 + [1100.1 \cdot (t - 24) + 0.9734 \cdot (t - 24)^2] \times 10^{-6}$$

The volume at 25° C was again determined June 10, 1910, and the value found to be 47.7186 ml. For determinations of density where the absolute value is sought, as in part 3 of this work, the mean value for this period, i. e., 47.71844 ml at 25° C is chosen.

4. BALANCE, WEIGHTS, AND METHOD OF WEIGHING

The balance used was a Rueprecht analytical balance of 200 g capacity, provided with special mechanism for rapid weighing and with the outside control of the weights of less than 1 g. The sensibility when undamped was 0.16 mg per division. The weights from 500 mg to 10 mg were the special weights belonging to the balance. The other weights used comprised a set of platinum-plated brass weights from 200 g to 1 g designated as B. S. No. 5157.

Previous to the experimental work the weights were carefully adjusted. A calibration made April, 1909, showed that the error of any possible combination did not exceed +0.08 mg.

A retest made June, 1910, after the completion of the investigation showed the maximum error of any combination to be +0.23 mg. This change is greater than was anticipated, yet is sufficiently small to be disregarded.

The weighings of the sinker in the liquid were made by the method of substitution. A constant counterpoise was kept on the left pan of the balance, while the known weights were applied on the right, from which the sinker was suspended. Equilibrium was first obtained with the immersed sinker attached, then the sinker was detached and left resting on the bottom of the tube while equilibrium was again obtained with the suspending hook alone. The difference in the weights required to secure equilibrium is the apparent weight of the sinker in the liquid.

The suspension wire where it passed through the surface of the liquid was of platinum 0.3 mm in diameter. It was covered with a layer of dull gold by electrodeposition in order to insure wetting with the liquid, and thus avoid the sticking otherwise produced. With the wire thus prepared, weighings could be made to 0.1 mg when the sinker was not attached and to about 0.3 mg with the sinker attached. Since the accidental error of weighing is subject to elimination in the final reduction, weighings to 0.5 mg were accepted as being comparable with the precision in other elements of the work.

All weighings were reduced to vacuo by means of a buoyancy balance, which was devised for obtaining the buoyancy correction to weighings of water for volumetric determinations. This apparatus consists of a hollow bulb of glass having an external volume of 900 cc, suspended from one arm of a balance and counterpoised by a brass weight of equal mass. The weight of air equal in volume to the difference in volume of these two objects is indicated by the weights required on the bulb side to secure a balance. The difference in volume is 881.3 cc; thus the buoyancy on 1 liter of water weighed with brass weights is obtained directly. The air density is obtained by multiplying this observed "buoyancy constant" by $\frac{1}{881.3}$. The bulb is suspended in a glass case to protect it from disturbing air currents. Correction is made for difference in temperature between this case and the balance where the

determinations are made. The density of the weights for the purpose of correcting for displaced air is assumed to be 8.4.

5. MATERIAL USED

The water used in the preparation of the mixtures investigated was twice distilled, the second distillation being from alkaline potassium permanganate. The three samples of alcohol used were all prepared from sample A, previously described. The densities at 4°C and corresponding percentages of alcohol are given below:

Sample	$D_{4^{\circ}\text{C}}^{25^{\circ}}$	Per cent alcohol by weight
L_3	0.78529	99.93
$3\ b$.78533	99.91
I_3	.78507	100.00

Tests made by the chemical division of the bureau failed to show the presence of impurities other than water in sufficient amounts to affect the density appreciably, and it may be safely assumed that the effect on the thermal expansion of these last traces of impurities is less than the experimental error of the determinations.

6. ARRANGEMENT OF EXPERIMENTAL WORK

In order to make the final results of the determination of the thermal expansion as free as possible from the effects of progressive changes in density that are not directly attributable to changes in temperature, the observations were arranged in two series, with the order of the temperatures reversed, the temperatures of density determinations being as follows:

First series	10	15	20	25	30	35	40
Second series	40	35	30	25	20	15	10

In each series the observations were distributed at approximately equal time intervals. Each series required a day for completion.

By employing this isochronously symmetrical arrangement of observations and by taking the mean of the two series disturbing causes which are linear functions of time are practically eliminated.

The principal disturbing causes which were anticipated were evaporation of alcohol and absorption of air and moisture.

Notwithstanding the precautions for the exclusion of atmospheric moisture and prevention of evaporation from the densimeter tube when not actually observing, the change in concentration due to these causes was found to be appreciable in most of the mixtures. Owing to the lower vapor pressure of alcohol the vapor usually is of higher concentration than the mixture which yields it, and thus a progressive dilution of the mixture is produced in most cases. That this dilution is uniformly distributed throughout the mixture is unlikely. There is besides, with some of the mixtures, condensation of the vapor in the top of the tube and on the suspension, which tends to run back into the upper layers of the liquid. This may account for the fact that in the 90 per cent mixture there appeared to be between the series an increase in concentration. The increase in the concentration of the 10 per cent mixture, however, was artificial, being caused by the intentional addition of alcohol to compensate for evaporation.

It was found that unless dissolved air was extracted from the mixtures there was danger of air being expelled from the liquid at the higher temperatures and attaching to the sinker as bubbles, preventing a successful series of observations. Hence, before the determination for any mixture, and in some cases between the two series of observations, the mixture was evacuated to low pressure to remove a sufficient amount of the dissolved air to prevent this difficulty. During the series air was doubtless reabsorbed and at an unknown rate, but determinations made elsewhere of the effect of dissolved air on the density of various mixtures at 25° C showed the total effect at that temperature to be so small that any large variable effect at the temperatures employed and in the time allowed would be improbable.

Examination of the experimental results shows that the alteration in concentration of the mixtures was usually less for the higher than for the lower per cents of alcohol. It is also seen that between the first and second series of observations on several of the mixtures a considerable change occurred. This can be attributed to the evacuation of the mixture previous to the second series to remove air by which alcohol vapor was also removed.

The manner of observing was as follows: With the liquid to be investigated in the tube, the sinker immersed, the cap on and closed by the suspension plug, and the tube in place in the control bath, the temperature was brought to the initial temperature of the series. The brine flow, the by-pass in the circulating apparatus, the thermoregulator, and the electric energy supply were successively adjusted to maintain the desired constant temperature. Sufficient time was allowed for the inner bath to acquire a constant or very slowly rising temperature. The sinker was then suspended from the balance and weighed. The thermometers were both read. The sinker was detached and the suspension weighed. The sinker was again suspended and weighed and the thermometers again read. At the conclusion of this set of observations the top of the densimeter tube was closed by the suspension plug and the temperature changed to the next one of the series as rapidly as the capacity of the heating coil or of the cooling coil would permit. While this temperature change was taking place the ice points of the thermometers were read and the buoyancy constant observed. The temperature control was readjusted for constant regulation at the new temperature and observations for density and temperature were again made. This procedure was repeated until the series was complete.

V. EXPERIMENTAL RESULTS

1. CALCULATION OF RESULTS

The total correction to the thermometer at the temperature of the determination was obtained by use of the observed ice points and the calibration corrections. Each observed temperature was corrected and the mean of the set of four taken as the temperature of the determination.

The apparent weight of the sinker in the liquid is given by the difference between the balance readings with sinker on and with sinker off. The mean of the two values of the apparent weight of sinker thus obtained is corrected by subtracting the weight of air displaced by the weights. To obtain this correction the volume of the weights (assuming their density to be 8.4) is multiplied by the observed air density. The application of this correction gives the true weight of the sinker in the liquid. The difference between

TABLE XXIII
Determination of Thermal Expansion of Alcohol-Water Mixture Containing 49.961 Per Cent Alcohol
Record of Observations and Calculations

SERIES I.—JAN. 28, 1910. RECORD: VOL. 646, p. 122. OBSERVERS: N. S. O., H. W. B.

Time	Observed temperature		Observed ice point and <i>Total correction</i>		Corrected temperature		Mean temperature of set <i>t'</i>	Balance reading (a) Sinker on (b) <i>Sinker off</i> (c) Sinker on	Apparent weight of sinker in liquid	Buoyancy correction on weights	True weight of sinker in liquid	Weight of displaced liquid	Volume of sinker at <i>t</i> °	Density at observed temperature $D_{\frac{t}{4}}$
	Haak No. 2040	Tonnclot No. 4653	Haak No. 2040	Tonnclot No. 4653	Haak No. 2040	Tonnclot No. 4653								
a. m.														
11.07	10.020	9.925	+0.088	−0.100	10.019	10.016	39.2972	g	56.0427	−0.0078	56.0349	43.9643	47.70221	0.921641
11.12							95.3399						
11.14	10.025	9.927	−0.001	+0.091	10.024	10.018	39.2972		56.0427					
p. m.														
12.14	15.020	14.940	+0.085	−0.102	15.012	15.005	39.1120		56.2213	−0.0078	56.2136	43.7856	47.70759	0.917791
12.18	15.025	14.945	−0.008	+0.065	15.017	15.010	95.3333						
12.21							39.1118		56.2215					
1.16	19.995	19.945	+0.082	−0.110	20.000	19.986	38.9238		56.4027	−0.0079	56.3949	43.6043	47.71301	0.913887
1.19							95.3265						
1.24	19.990	19.950	+0.005	+0.041	19.995	19.991	38.9236		56.4029					
2.05	25.015	24.980	+0.080	−0.115	25.005	25.003	38.7314		56.5882	−0.0079	56.5804	43.4188	47.71851	0.909894
2.07							95.3196						
2.11	25.020	24.985	−0.010	+0.023	25.010	25.008	38.7312		56.5884					
2.49	29.980	29.970	+0.075	−0.118	29.985	29.983	38.5371		56.7757	−0.0079	56.7678	43.2314	47.72403	0.905862
2.54							95.3128						
2.57	29.985	29.973	+0.005	+0.013	29.990	29.986	38.5371		56.7757					
	Richter No. 264		Richter No. 264		Richter No. 264									
3.30	34.987	34.925	+0.027	−0.120	34.950	34.960	38.3404		56.9652	−0.0079	56.9571	43.0421	47.72958	0.901791
3.33							95.3056						
3.38	34.998	34.930	−0.037	+0.035	34.961	34.965	38.3407		56.9649					
4.13	39.990	39.880	+0.025	−0.125	39.957	39.958	38.1386		57.1597	−0.0080	57.1517	42.8475	47.73521	0.897608
4.17							95.2983						
4.20	39.995	39.880	−0.033	+0.078	39.962	39.958	38.1386		57.1597					

Air temperature: In balance, 21° 5; at buoyancy bulb, 19° 6. Buoyancy constant: Observed, 1040; corrected, 1032. Air density, 0.001172 g/cm³. Weight of sinker, 99.9990 g.

the weight of the sinker in the liquid and its weight in vacuo gives the weight of the liquid displaced by the sinker. This weight divided by the volume of the sinker at the temperature of the determination gives the density of the liquid at that temperature in grams per milliliter.

The calculation of the density of the liquid as described above may be expressed by the following equation:

$$D_{\frac{t}{4}} = \frac{S - \frac{W_1 - w + W_2 - w}{2} \left(1 - \frac{\rho}{8.4}\right)}{V_t}$$

Where $D_{\frac{t}{4}}$ = density of sample at temperature t

S = mass of sinker

V_t = volume of sinker at the temperature t

t = temperature of determination

W_1, W_2 = balance readings with sinker on

w = balance readings with sinker off

ρ = air density

2. COPY OF ORIGINAL DATA

A copy of the record showing the observations comprising the two series of determinations for a single mixture is given in Table XXIII (pp. 382-383). This table illustrates the arrangement of the experimental work and also the calculation of results as described above.

3. REDUCTION AND ADJUSTMENT OF RESULTS

From the observations and calculations as described and illustrated above are obtained the experimental results given in Tables XXIV to XXXV, comprising for each mixture a double series of density determinations together with the corresponding temperatures. These temperatures are all very close to the integral temperatures intended. For the purpose of simplifying the adjustment of results the determined densities are reduced to the values corresponding to the integral temperatures. Comparison between the densities thus reduced shows the permanent change occurring in the liquid during the progress of the determination not directly attributable to the change in temperature. This permanent change which is caused by the evaporation of alcohol and by the absorption of moisture may be regarded as a linear function of the position of the determination in the series, since the different groups of observations in a series occupy approximately the same time and are separated by equal intervals of time. The total permanent change occurring between any determination of the first series and the corresponding one of the second is represented by the equation:

$$\Delta = a + b (7 - n)$$

where n is the number of the determination in the first series, a is the change occurring between the first and second series—that is, between the seventh and eighth determinations—and may include changes from any intentional or accidental cause, b depends on the rate of change during the progress of a single series and is assumed to be the same in the two series. By a least square adjustment of the observed apparent permanent change in density between the series, the constants a and b are determined. The adjusted value of Δ is then calculated by these coefficients, and each determined density corrected by one-half the corresponding adjusted Δ with the appropriate sign.

It is evident that the final value of the density at each temperature will be the same whether the mean be taken of the two determinations at that temperature or whether these determinations be first adjusted, as indicated above, to correct for the progressive change of density due to evaporation of alcohol and

absorption of water vapor. This adjustment, however, is desirable in order that a check may be had upon the agreement of the two determinations when all explainable differences have been taken into account. This agreement is an indication of the magnitude of the accidental errors of the individual determinations.

The observed densities thus adjusted to values corresponding to the mean concentration of the mixture are further adjusted on the assumption that the equation

$$D_4^t = D_4^{25} + \alpha (t - 25) + \beta (t - 25)^2 + \gamma (t - 25)^3$$

represents the change in density depending on the change in temperature alone. By the method of least squares, $D_4^{25}, \alpha, \beta, \gamma$ are determined. The adjusted value of D_4^t is then calculated. In the values of observed D_4^t minus calculated D_4^t will be combined the accidental experimental errors as well as errors due to the assumed form of functions in the least square adjustments of the observations. It appears from examination of these residuals that the assumptions made are in accordance with the experimental data, unless these errors are systematically compensated, as the residuals are seen to be within the limits of accidental error.

TABLE XXIV

Determination of Thermal Expansion of Alcohol-Water Mixture Containing 4.907 Per Cent Alcohol by Weight
Reduction and Adjustment of Experimental Results

Determina- tion number	Observed temperature t'	Observed density $D_4^{t'}$	Reduction to integral temperature		Apparent permanent change between series $\Delta' \times 10^6$	Adjusted correction for permanent change $\frac{\Delta}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	$D_4^{t'}$						
1	39.931	0.983244	40	0.983216	100	+49	0.983265	0.983266	0.983267	-2
14	39.930	.983344		.983316		-49	.983267			0
2	34.984	.985125	35	.985119	96	+48	.985167	.985167	.985163	+4
13	34.963	.985228		.985215		-48	.985167			+4
3	30.021	.986800	30	.986807	92	+48	.986855	.986853	.986854	+1
12	30.017	.986894		.986899		-48	.986851			-3
4	25.028	.988262	25	.988270	84	+47	.988317	.988312	.988317	0
11	25.042	.988343		.988354		-47	.988307			-10
5	20.008	.989478	20	.989480	107	+46	.989526	.989534	.989530	-4
10	20.018	.989583		.989587		-46	.989541			+11
6	15.018	.990419	15	.990422	92	+46	.990468	.990468	.990468	0
9	15.022	.990511		.990514		-46	.990468			0
7	10.053	.991060	10	.991065	86	+45	.991110	.991108	.991108	+2
8	10.036	.991147		.991151		-45	.991106			-2

Equation for adjustment of Δ : $\Delta = 90 + 1.285(7 - n)$.

Probable value of D_4^t from above determinations: $D_4^t = 0.988317 - [268.4(t - 25) + 5.02(t - 25)^2 - 0.0311(t - 25)^3]10^6$.

TABLE XXV
Determination of Thermal Expansion of Alcohol-Water Mixture Containing 9.984 Per Cent of Alcohol by Weight
Reduction and Adjustment of Experimental Results

Determina- tion number	Observed temperature t'	Observed density $D_4^{t'}$	Reduction to integral temperature		Apparent permanent change between series $A' \times 10^6$	Adjusted correction for permanent change $\frac{A}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calulated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	$D_4^{t'}$						
1	40.002	0.974768	40	0.974769	22	+ 8	0.974777	0.974780	0.974781	-4
14	39.966	.974806		.974791		-8	.974783			+2
2	35.001	.976880	35	.976880	11	+6	.976886	.976886	.976884	+2
13	34.976	.976901		.976891		-6	.976885			+1
3	30.026	.978774	30	.978783	1	+4	.978787	.978784	.978784	+3
12	29.989	.978788		.978784		-4	.978780			-4
4	25.016	.980458	25	.980463	-5	+2	.980465	.980460	.980461	+4
11	25.019	.980452		.980458		-2	.980456			-5
5	20.019	.981887	20	.981892	5	0	.981892	.981894	.981896	-4
10	20.022	.981891		.981897		0	.981897			+1
6	15.038	.983063	15	.983071	5	-2	.983069	.983074	.983070	-1
9	15.020	.983072		.983076		+2	.983078			+8
7	10.039	.983962	10	.983968	-11	-4	.983964	.983962	.983963	+1
8	10.048	.983950		.983957		+4	.983961			-2

Equation for adjustment of A : $A = -7.5 + 3.822 (7 - n)$.
Probable value of D_4^t from above determinations: $D_4^t = 0.980461 - [0.311.9 (t - 25) + 4.84 (t - 25)^2 - 0.0258 (t - 25)^3] 10^{-6}$.

TABLE XXVI
Determination of Thermal Expansion of Alcohol-Water Mixture Containing 19.122 Per Cent Alcohol by Weight
Reduction and Adjustment of Experimental Results

Determina- tion number	Observed temperature t'	Observed density $D_4^{t'}$	Reduction to integral temperature		Apparent permanent change between series $d' \times 10^6$	Adjusted correction for permanent change $\frac{d}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	D_4^t						
1	10. 052	0. 973455	10	0. 973472	35	+22	0. 973494	0. 973490	0. 973492	+ 2
14	10. 037	. 973495		. 973507						
2	15. 021	. 971736	15	. 971744	42	+20	. 971764	. 971765	. 971763	+ 1
13	15. 028	. 971776		. 971786						
3	20. 025	. 969783	20	. 969793	37	+19	. 969812	. 969812	. 969810	+ 2
12	20. 033	. 969816		. 969830						
4	25. 020	. 967608	25	. 967617	51	+18	. 967635	. 967642	. 967648	-13
11	25. 042	. 967649		. 967668						
5	29. 992	. 965275	30	. 965271	37	+17	. 965288	. 965290	. 965289	- 1
10	30. 036	. 965290		. 965308						
6	34. 990	. 962742	35	. 962737	26	+16	. 962753	. 962750	. 962747	+ 6
9	34. 993	. 962767		. 962763						
7	39. 966	. 960042	40	. 960023	23	+14	. 960037	. 960034	. 960036	+ 1
8	39. 966	. 960065		. 960046						

Equation for adjustment of d : $d = 28.6 + 2.430 (7 - n)$.
Probable value of D_4^t from above determinations: $D_4^t = 0.967648 - [452.6 (t - 25) + 3.93 (t - 25)^2 - 0.0180 (t - 25)^3] 10^{-6}$.

TABLE XXVII
Determination of Thermal Expansion of Alcohol-Water Mixture Containing 22.918 Per Cent Alcohol by Weight
Reduction and Adjustment of Experimental Results

Determina- tion number	Observed temperature t	Observed density D_4^t	Reduction to integral temperature		Apparent permanent change $d' \times 10^6$	Adjusted correction for permanent change $\frac{d}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	$D_4^{t'}$						
1	10. 015	0. 969152	10	0. 969158	64	+38	0. 969196	0. 969190	0. 969190	+ 6
14	10. 021	. 969213		. 969222		-38	. 969184			- 6
2	14. 995	. 966981	15	. 966979	71	+36	. 967015	. 967014	. 967016	- 1
13	15. 011	. 967045		. 967050		-36	. 967014			- 2
3	20. 002	. 964626	20	. 964627	73	+35	. 964662	. 964664	. 964660	+ 2
12	20. 011	. 964695		. 964700		-35	. 964665			+ 5
4	24. 989	. 962097	25	. 962091	76	+33	. 962124	. 962129	. 962133	- 9
11	25. 019	. 962157		. 962167		-33	. 962134			+ 1
5	29. 990	. 959408	30	. 959402	76	+32	. 959434	. 959440	. 959440	- 6
10	29. 998	. 959479		. 959478		-32	. 959446			+ 6
6	34. 981	. 956568	35	. 956557	64	+30	. 956587	. 956589	. 956588	- 1
9	34. 980	. 956633		. 956621		-30	. 956591			+ 3
7	39. 972	. 953583	40	. 953566	40	+29	. 953595	. 953586	. 953587	+ 8
8	39. 955	. 953634		. 953606		-29	. 953577			-10

Equation for adjustment of d : $d = 57.4 + 2.963(7 - n)$.
Probable value of D_4^t from above determinations: $D_4^t = 0.962133 - [522.4(t - 25) + 3.31(t - 25)^2 - 0.0100(t - 25)^3] 10^{-6}$.

TABLE XXVIII

Determination of Thermal Expansion of Alcohol-Water Mixture Containing 30.086 Per Cent Alcohol, by Weight

Osborne]

Density and Expansion of Alcohol

Reduction and Adjustment of Experimental Results

Determina- tion number	Observed temperature t'	Observed density $D_4^{t'}$	Reduction to integral temperature		Apparent permanent change between series $d' \times 10^6$	Adjusted correction for permanent change $\frac{d}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^4$
			t	$D_4^{t'}$						
1	10. 024	0. 959538	10	0. 959552	201	+103	0. 959655	0. 959652	0. 959650	+ 5
14	10. 019	. 959742		. 959753		-103	. 959650			0
2	15. 001	. 956626	15	. 956627	194	+102	. 956729	. 956724	. 956729	0
13	15. 011	. 956814		. 956821		-102	. 956719			-10
3	20. 002	. 953589	20	. 953590	205	+100	. 953690	. 953692	. 953686	+ 4
12	20. 014	. 953786		. 953795		-100	. 953695			+ 9
4	25. 011	. 950422	25	. 950429	199	+ 99	. 950528	. 950528	. 950529	- 1
11	25. 022	. 950614		. 950628		- 99	. 950529			0
5	29. 986	. 947160	30	. 947151	216	+ 97	. 947248	. 947259	. 947258	-10
10	30. 006	. 947363		. 947367		- 97	. 947270			+12
6	34. 964	. 943796	35	. 943771	206	+ 96	. 943867	. 943874	. 943877	-10
9	34. 983	. 943989		. 943977		- 96	. 943881			+ 4
7	39. 963	. 940335	40	. 940309	163	+ 95	. 940404	. 940390	. 940390	+14
8	39. 958	. 940501		. 940472		- 95	. 940377			-13

Equation for adjustment of d : $d = 189.2 + 2.841(7 - n)$.
Probable value of D_4^t from above determinations: $D_4^t = 0.950529 - [643.1(t - 25) + 2.26(t - 25)^2 - 0.0047(t - 25)^3]10^{-6}$.

TABLE XXIX
Determination of Thermal Expansion of Alcohol-Water Mixture Containing 39.988 Per Cent Alcohol, by Weight
Reduction and Adjustment of Experimental Results

Determina- tion number	Observed temperature t'	Observed density $D_4^{t'}$	Reduction to integral temperature		Apparent permanent change $\Delta' \times 10^6$ between series	Adjusted correction for permanent change $\frac{\Delta}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	$D_4^{t'}$						
1	10. 017	0. 942340	10	0. 942352	126	+68	0. 942420	0. 942415	0. 942415	+5
14	10. 035	. 942453		. 942478		-68	. 942410			-5
2	15. 014	. 938777	15	. 938787	128	+65	. 938852	. 938851	. 938851	+1
13	15. 020	. 938901		. 938915		-65	. 938850			-1
3	20. 007	. 935149	20	. 935154	130	+63	. 935217	. 935219	. 935215	+2
12	20. 018	. 935271		. 935284		-63	. 935221			+6
4	25. 012	. 931431	25	. 931440	124	+60	. 931500	. 931502	. 931507	-7
11	25. 020	. 931564		. 931564		-60	. 931504			-3
5	29. 984	. 927680	30	. 927668	118	+57	. 927725	. 927727	. 927727.	-2
10	29. 997	. 927788		. 927786		-57	. 927729			+2
6	34. 998	. 923818	35	. 923816	119	+55	. 923871	. 923876	. 923874	-3
9	34. 982	. 923949		. 923935		-55	. 923880			+6
7	39. 946	. 919942	40	. 919899	95	+52	. 919951	. 919946	. 919947	+4
8	39. 969	. 920019		. 919994		-52	. 919942			-5

Equation for adjustment of Δ : $\Delta = 103.6 + 5.464 (7 - n)$.
Probable value of D_4^t from above determinations: $D_4^t = 0.931507 - [748.8(t - 25) + 1.45(t - 25)^2 + 0.0004(t - 25)^3]10^{-6}$.

TABLE XXX

Determination of Thermal Expansion of Alcohol-Water Mixture Containing 49.961 Per Cent Alcohol by Weight
Reduction and Adjustment of Experimental Results

Determination number	Observed temperature t'	Observed density $D_4^{t'}$	Reduction to integral temperature		Apparent permanent change between series $d' \times 10^6$	Adjusted correction for permanent change $\frac{d}{2} \times 10^4$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	$D_4^{t'}$						
1	10. 019	0. 921641	10	0. 921655	99	+52	0. 921707	0. 921704	0. 921705	+ 2
14	10. 020	. 921739		. 921754		-52	. 921702			- 3
2	15. 011	. 917791	15	. 917800	94	+47	. 917847	. 917847	. 917844	+ 3
13	15. 014	. 917883		. 917894		-47	. 917847			+ 3
3	19. 993	. 913887	20	. 913881	83	+41	. 913922	. 913922	. 913922	0
12	20. 003	. 913962		. 913964		-41	. 913923			+ 1
4	25. 006	. 909894	25	. 909899	78	+35	. 909934	. 909938	. 909937	- 3
11	25. 015	. 909965		. 909977		-35	. 909942			+ 5
5	29. 986	. 905862	30	. 905851	59	+30	. 905881	. 905880	. 905888	- 7
10	29. 998	. 905912		. 905910		-30	. 905880			- 8
6	34. 959	. 901791	35	. 901757	53	+24	. 901781	. 901784	. 901774	+ 7
9	34. 986	. 901822		. 901810		-24	. 901786			+12
7	39. 959	. 897608	40	. 897573	30	+19	. 897592	. 897588	. 897591	+ 1
8	39. 954	. 897642		. 897603		-19	. 897584			- 7

Equation for adjustment of d : $d = 37.5 + 11.14(7 - n)$.
 Probable value of D_4^t from above determinations: $D_4^t = 0.909937 - [803.3(t - 25) + 1.28(t - 25)^2 + 0.0024(-25)^3]10^{-6}$.

TABLE XXXI
Determination of Thermal Expansion of Alcohol-Water Mixture Containing 59.976 Per Cent Alcohol by Weight
Reduction and Adjustment of Experimental Results

Determination number	Observed temperature t'	Observed density $D_4^{t'}$	Reduction to integral temperature		Apparent permanent change between series $d' \times 10^6$	Adjusted correction for permanent change $\frac{d}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	$D_4^{t'}$						
1	10. 023	0. 899275	10	0. 899293	60	+33	0. 899326	0. 899323	0. 899323	+3
14	10. 024	. 899334		. 899353		-33	. 899320			-3
2	14. 998	. 895260	15	. 895258	62	+31	. 895289	. 895289	. 895290	-1
13	15. 025	. 895300		. 895320		-31	. 895289			-1
3	20. 001	. 891172	20	. 891173	57	+29	. 891202	. 891202	. 891200	+2
12	20. 028	. 891230		. 891230		-29	. 891201			+1
4	25. 018	. 887002	25	. 887017	64	+27	. 887044	. 887049	. 887051	-7
11	25. 043	. 887045		. 887081		-27	. 887054			+3
5	29. 980	. 882832	30	. 882815	54	+26	. 882841	. 882842	. 882842	-1
10	29. 996	. 882872		. 882869		-26	. 882843			+1
6	34. 974	. 878566	35	. 878544	51	+24	. 878568	. 878570	. 878570	-2
9	34. 976	. 878616		. 878595		-24	. 878571			+1
7	39. 969	. 874243	40	. 874216	34	+22	. 874238	. 874233	. 874233	+5
8	39. 977	. 874220		. 874250		-22	. 874228			-5

Equation for adjustment of d : $d = 43.6 + 3.68(7 - n)$.
Probable value of D_4^t from above determinations: $D_4^t = 0.887051 - [835.8(t - 25) + 1.21(t - 25)^2 + 0.0024(t - 25)^3]10^{-6}$.

TABLE XXXII

Determination of Thermal Expansion of Alcohol-Water Mixture Containing 70.012 Per Cent Alcohol by Weight
Reduction and Adjustment of Experimental Results

Determina- tion number	Observed temperature t'	Observed density $D_4^{t'}$	Reduction to integral temperature		Apparent permanent change between series $d' \times 10^6$	Adjusted correction for permanent change $\frac{d}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	$D_4^{t'}$						
1	10. 034	0. 875938	10	0. 875965	46	+21	0. 875987	0. 875989	0. 875989	-2
14	10. 018	. 875997		. 876012		-21	. 875991			+2
2	15. 014	. 871819	15	. 871831	34	+20	. 871851	. 871848	. 871845	+6
13	15. 023	. 871846		. 871865	"	-20	. 871845			0
3	20. 002	. 867621	20	. 867623	33	+19	. 867642	. 867640	. 867641	+1
12	20. 022	. 867637		. 867656		-19	. 867637			-4
4	25. 016	. 863344	25	. 863358	39	+18	. 863376	. 863378	. 863380	-4
11	25. 040	. 863363		. 863397		-18	. 863379			-1
5	29. 995	. 859045	30	. 859041	38	+16	. 859057	. 859060	. 859060	-3
10	30. 004	. 859076		. 859079		-16	. 859063			+3
6	34. 988	. 854675	35	. 854664	38	+15	. 854679	. 854683	. 854681	-2
9	34. 998	. 854704		. 854702		-15	. 854687			+6
7	39. 979	. 850250	40	. 850231	19	+14	. 850245	. 850240	. 850241	+4
8	39. 988	. 850261		. 850250		-14	. 850236			-5

Equation for adjustment of d : $d = 28.0 + 2.428 (7 - n)$.
Probable value of D_4^t from above determinations: $D_4^t = 0.863380 - [858.1 (t - 25) + 1.17 (t - 25)^2 + 0.0009 (t - 25)^3] 10^{-6}$.

TABLE XXXIII
Determination of Thermal Expansion of Alcohol-Water Mixture Containing 80.036 Per Cent Alcohol by Weight
Reduction and Adjustment of Experimental Results

Determina- tion number	Observed temperature t'	Observed density $D_4^{t'}$	Reduction to integral temperature		Apparent permanent change $d' \times 10^6$ between series	Adjusted correction for permanent change $\frac{d}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	$D_4^{t'}$						
1	10. 032	0. 851856	10	0. 851883	- 1	- 1	0. 851882	0. 851882	0. 851882	0
14	10. 026	. 851860		. 851882	+ 1	+ 1	. 851883			+1
2	15. 028	. 847619	15	. 847643	- 1	0	. 847643	. 847642	. 847644	-1
13	15. 019	. 847626		. 847642		0	. 847642			-2
3	20. 025	. 843342	20	. 843364	- 2	+ 1	. 843365	. 843363	. 843362	+3
12	20. 029	. 843337		. 843362		- 1	. 843361			-1
4	25. 034	. 838997	25	. 839027	5	+ 2	. 839029	. 839030	. 839031	-2
11	25. 025	. 839010		. 839032		- 2	. 839030			-1
5.	30. 006	. 834636	30	. 834641	11	+ 3	. 834644	. 834646	. 834646	-2
10	30. 020	. 834634		. 834652		- 3	. 834649			+3
6	34. 988	. 830210	35	. 830199	6	+ 4	. 830203	. 830202	. 830202	+1
9	34. 994	. 830210		. 830205		- 4	. 830201			-1
7	39. 966	. 825722	40	. 825691	7	+ 5	. 825696	. 825694	. 825694	+2
8	39. 981	. 825715		. 825698		- 5	. 825693			-1

Equation for adjustment of d : $d = 9.1 - 1.821 (7 - n)$.
Probable value of D_4^t from above determinations: $D_4^t = 0.839031 - [871.4 (t - 25) + 1.08 (t - 25)^2 + 0.0069 (t - 25)^3] 10^{-6}$.

TABLE XXXIV
Determination of Thermal Expansion of Alcohol-Water Mixture Containing 90.037 Per Cent Alcohol by Weight
Reduction and Adjustment of Experimental Results

Determina- tion number	Observed temperature t'	Observed density D'_4	Reduction to integral temperature		Apparent permanent change between series $d' \times 10^6$	Adjusted correction for permanent change $\frac{d}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	D''_4^t						
1	10. 029	0. 826431	10	0. 826456	-29	-15	0. 826441	0. 826442	0. 826443	-2
14	10. 034	. 826398		. 826427		+15	. 826442			-1
2	15. 022	. 822169	15	. 822188	-28	-14	. 822174	. 822174	. 822174	0
13	15. 031	. 822133		. 822160		+14	. 822174			0
3	20. 015	. 817862	20	. 817875	-18	-12	. 817863	. 817866	. 817866	-3
12	20. 010	. 817848		. 817857		+12	. 817869			+3
4	25. 012	. 813519	25	. 813529	-28	-11	. 813518	. 813515	. 813516	+2
11	25. 036	. 813470		. 813501		+11	. 813512			-4
5	29. 986	. 809144	30	. 809132	-25	-10	. 809122	. 809120	. 809120	+2
10	30. 009	. 809099		. 809107		+10	. 809117			-3
6	34. 980	. 804698	35	. 804680	-16	-9	. 804671	. 804672	. 804673	-2
9	34. 996	. 804668		. 804664		+9	. 804673			0
7	39. 990	. 800187	40	. 800178	-14	-8	. 800170	. 800171	. 800172	-2
8	39. 987	. 800176		. 800164		+8	. 800172			0

Equation for adjustment of d : $d = -16 - 2.213 (7 - n)$.
Probable value of D_4^t from above determinations: $D_4^t = 0.813516 - [874.6 (t - 25) + 0.93 (t - 25)^2 + 0.0051 (t - 25)^3] 10^{-6}$.

TABLE XXXV
Determination of Expansion of Alcohol-Water Mixture Containing 99.913 Per Cent Alcohol by Weight
Reduction and Adjustment of Experimental Results

Determination number	Observed temperature t'	Observed density $D_4^{t'}$	Reduction to integral temperature		Apparent permanent change between series $d' \times 10^6$	Adjusted correction for permanent change $\frac{d}{2} \times 10^6$	Density adjusted to mean concentration D_4^t	Mean observed density D_4^t	Calculated D_4^t	Observed D_4^t minus calculated $D_4^t \times 10^6$
			t	$D_4^{t'}$						
1	10.036	0.798080	10	0.798110	13	+8	0.798118	0.798116	0.798118	0
14	10.018	.798108		.798123		-8	.798115			-3
2	15.005	.793868	15	.793872	21	+8	.793880	.793882	.793879	+1
13	15.022	.793874		.793893		-8	.793885			+6
3	19.999	.789610	20	.789609	19	+8	.789617	.789618	.789620	-3
12	20.031	.789602		.789628		-8	.789620			0
4	25.022	.785307	25	.785326	17	+8	.785334	.785334	.785337	-3
11	25.033	.785315		.785343		-8	.785335			-2
5	30.008	.781015	30	.781022	10	+8	.781030	.781027	.781026	+4
10	30.010	.781023		.781032		-8	.781024			-2
6	34.975	.776698	35	.776676	12	+8	.776684	.776682	.776681	+3
9	34.993	.776694		.776688		-8	.776680			-1
7	39.947	.772334	40	.772287	20	+8	.772295	.772297	.772298	-3
8	39.966	.772337		.772307		-8	.772299			+1

Equation for adjustment of d : $d = 15.4 + 0.214 (7 - n)$.
Probable value of D_4^t from above determinations: $D_4^t = 0.785337 - [859.3 (t - 25) + 0.57 (t - 25)^2 + 0.0062 (t - 25)^3] 10^{-6}$.

4. RÉSUMÉ OF RESULTS

By reduction and adjustment of experimental results there is derived for each mixture of alcohol and water investigated an equation for thermal expansion of the form:

$$D_4^t = D_4^{25} + \alpha(t - 25) + \beta(t - 25)^2 + \gamma(t - 25)^3$$

D_4^{25} and the coefficients α , β , γ , having been determined from the experimental data.

The constant D_4^{25} for each mixture represents the mean density at 25° in terms of the density of water at 4° as the unit and is used for obtaining the mean concentration of the mixture used. A table showing the relation between density at 25° and concentration is given on page 424.

The assembled results of the experimental work on thermal expansion of alcohol-water mixtures are given in Table XXXVI. Included in this table are the corresponding coefficients for water calculated from the experimental work of Chappuis¹¹³² using the densities at 10, 15, 20, 25, 30, 35, 40° C, upon which the present work is based. The coefficients α , β , γ , are shown graphically as functions of the per cent of alcohol in Fig. 9.

Interpolated values of α , β , γ obtained by the graphical method are given for each integral per cent of alcohol in Table XXXVIIa. The interpolation is carried to the number of figures shown for the purpose of avoiding arithmetic errors in the calculation of the final density Table XLIX in part 3. They should not be assumed correct to the last figure given. Furthermore, they can not be assumed as true except for the range of temperature used in their determination—viz, 10° C to 40° C.

In Table XXXVIIb are given the coefficients A , B , C in the equation for thermal expansion of the form:

$$V_t = V_{25}[1 + A(t - 25) + B(t - 25)^2 + C(t - 25)^3]$$

these coefficients having been calculated for each integral per cent of alcohol from the corresponding coefficients α , β , γ , given in Table XXXVIIa.

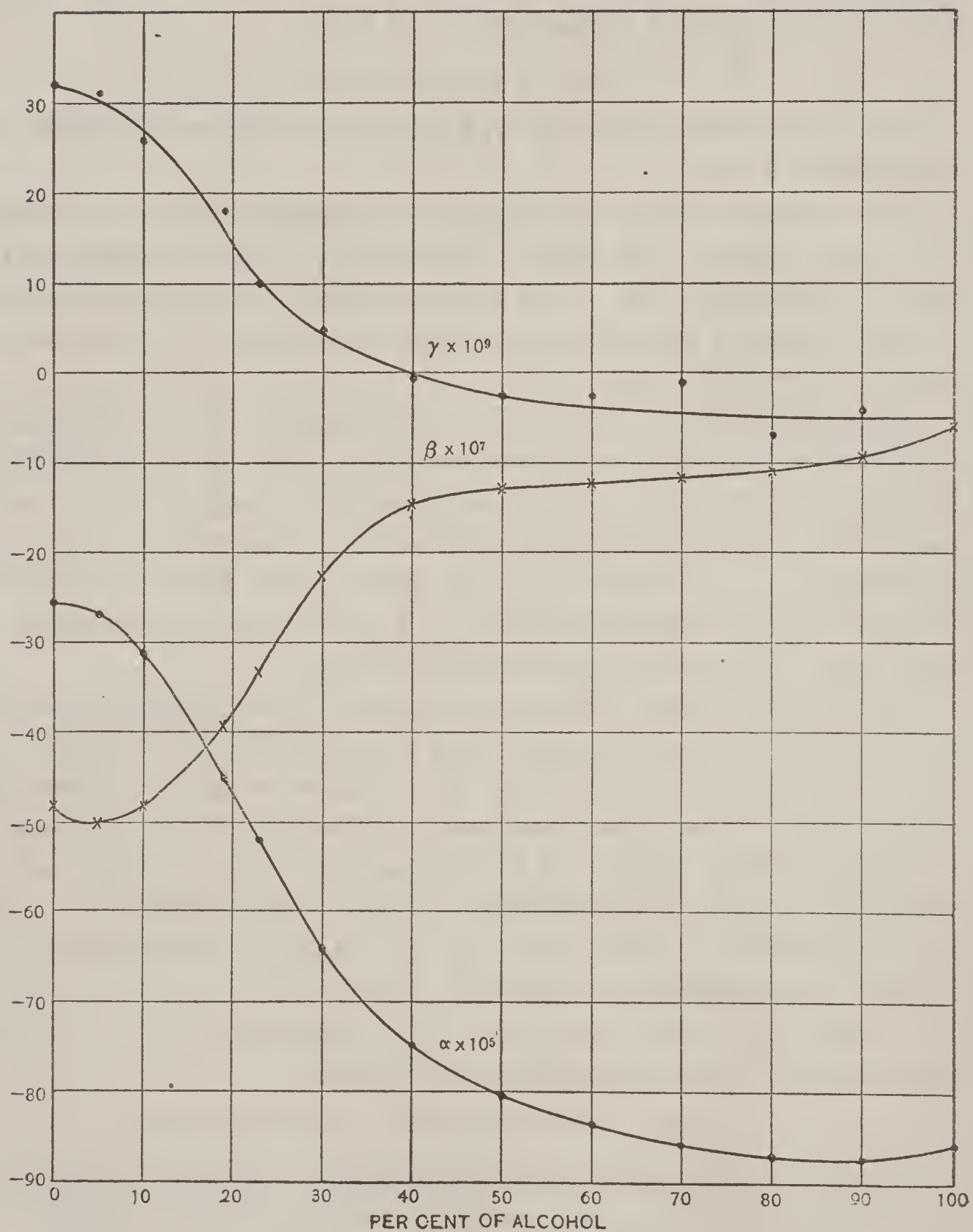
Fig. 9.—Coefficients α , β , γ

TABLE XXXVI

Thermal Expansion of Alcohol-Water Mixtures. Assembled Experimental Results

Equation: $D_4^t = D_4^{25} + \alpha(t - 25) + \beta(t - 25)^2 + \gamma(t - 25)^3$. Temperature range: 10° C to 40° C

Per cent alcohol by weight	D_4^{25}	$\alpha \times 10^7$	$\beta \times 10^8$	$\gamma \times 10^{10}$
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RESULTS FOR WATER ACCORDING TO CHAPPUIS'S DATA

0	0. 997077	−2565	−484	+319
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RESULTS OF PRESENT INVESTIGATION

4. 907	0. 988317	−2684	−502	+311
9. 984	. 980461	−3119	−484	+258
19. 122	. 967648	−4526	−393	+180
22. 918	. 962133	−5224	−331	+100
30. 086	. 950529	−6431	−226	+ 47
39. 988	. 931507	−7488	−145	− 4
49. 961	. 909937	−8033	−128	− 24
59. 976	. 887051	−8358	−121	− 24
70. 012	. 863380	−8581	−117	− 9
80. 036	. 839031	−8714	−108	− 69
90. 037	. 813516	−8746	− 93	− 51
99. 913	. 785337	−8593	− 57	− 62

TABLE XXXVIIa.—Interpolated values of α , β , γ , between 10°C to 40°C, in the equation:

$$D_4^t = D_4^{25} + \alpha(t - 25) + \beta(t - 25)^2 + \gamma(t - 25)^3$$

Per cent alcohol by weight	$\alpha \times 10^7$	$\beta \times 10^8$	$\gamma \times 10^9$	Per cent alcohol by weight	$\alpha \times 10^7$	$\beta \times 10^8$	$\gamma \times 10^9$
0	-2565	-484	+31.9	50	-8035	-128	-2
1	-2574	-490	+32	51	-8074	-127	-3
2	-2591	-496	+32	52	-8111	-126	-3
3	-2613	-499	+31	53	-8147	-126	-3
4	-2646	-501	+31	54	-8181	-125	-3
5	-2689	-502	+30	55	-8212	-124	-3
6	-2745	-501	+30	56	-8244	-124	-3
7	-2816	-498	+29	57	-8274	-123	-4
8	-2901	-495	+29	58	-8302	-122	-4
9	-3003	-490	+28	59	-8330	-122	-4
10	-3121	-484	+27	60	-8359	-121	-4
11	-3244	-476	+26	61	-8384	-121	-4
12	-3374	-469	+25	62	-8410	-120	-4
13	-3513	-460	+24	63	-8435	-120	-4
14	-3662	-452	+23	64	-8459	-120	-4
15	-3817	-442	+22	65	-8482	-119	-4
16	-3978	-432	+20	66	-8503	-119	-4
17	-4146	-420	+19	67	-8524	-119	-4
18	-4322	-408	+18	68	-8544	-118	-4
19	-4504	-395	+16	69	-8564	-117	-4
20	-4686	-380	+14	70	-8581	-117	-5
21	-4870	-363	+13	71	-8599	-116	-5
22	-5055	-346	+11	72	-8614	-116	-5
23	-5239	-329	+10	73	-8629	-115	-5
24	-5419	-313	+9	74	-8643	-114	-5
25	-5601	-298	+8	75	-8657	-113	-5
26	-5778	-282	+7	76	-8669	-112	-5
27	-5951	-268	+6	77	-8681	-111	-5
28	-6114	-253	+6	78	-8692	-110	-5
29	-6271	-240	+5	79	-8703	-109	-5
30	-6419	-227	+4	80	-8714	-108	-5
31	-6554	-215	+4	81	-8723	-107	-5
32	-6685	-204	+3	82	-8731	-106	-5
33	-6810	-194	+3	83	-8739	-105	-5
34	-6929	-185	+2	84	-8745	-104	-5
35	-7040	-176	+2	85	-8751	-102	-5
36	-7144	-168	+1	86	-8753	-101	-5
37	-7239	-161	+1	87	-8754	-99	-5
38	-7330	-155	+1	88	-8753	-97	-5
39	-7413	-150	0	89	-8751	-95	-5
40	-7489	-145	0	90	-8746	-93	-5
41	-7561	-141	0	91	-8738	-90	-5
42	-7627	-138	-1	92	-8728	-88	-5
43	-7689	-136	-1	93	-8715	-84	-5
44	-7748	-134	-1	94	-8700	-81	-5
45	-7802	-133	-1	95	-8685	-78	-5
46	-7855	-132	-2	96	-8668	-74	-5
47	-7903	-131	-2	97	-8650	-70	-5
48	-7950	-130	-2	98	-8632	-66	-5
49	-7993	-129	-2	99	-8613	-61	-5
50	-8035	-128	-2	100	-8591	-56	-5

TABLE XXXVIIb.—Values of A, B, C, between 10°C to 40°C, in the equation :

$$V_t = V_{25}[1 + A(t - 25) + B(t - 25)^2 + C(t - 25)^3]$$

Per cent alcohol by weight	A×10 ⁶	B×10 ⁷	C×10 ⁸	Per cent alcohol by weight	A×10 ⁶	B×10 ⁷	C×10 ⁸
0	+257.2	+49.2	−2.9	50	+ 883.1	+21.9	+0.5
1	+259	+50	−3	51	+ 890	+22	+1
2	+261	+51	−3	52	+ 896	+22	+1
3	+264	+51	−3	53	+ 902	+22	+1
4	+267	+51	−3	54	+ 908	+22	+1
5	+272.1	+51.5	−2.7	55	+ 914	+22	+1
6	+278	+52	−3	56	+ 920	+22	+1
7	+285	+51	−3	57	+ 926	+22	+1
8	+295	+51	−3	58	+ 931	+22	+1
9	+306	+51	−2	59	+ 937	+22	+1
10	+318.3	+50.4	−2.4	60	+ 942.4	+22.5	+0.7
11	+331	+50	−2	61	+ 948	+23	+1
12	+345	+49	−2	62	+ 953	+23	+1
13	+360	+48	−2	63	+ 958	+23	+1
14	+376	+48	−2	64	+ 964	+23	+1
15	+392	+47	−2	65	+ 969	+23	+1
16	+409	+46	−2	66	+ 974	+23	+1
17	+427	+45	−2	67	+ 979	+23	+1
18	+446	+44	−1	68	+ 984	+23	+1
19	+465.4	+43.0	−1.2	69	+ 989	+23	+1
20	+485	+42	−1	70	+ 993.9	+23.4	+0.9
21	+505	+40	−1	71	+ 999	+24	+1
22	+525	+39	−1	72	+1003	+24	+1
23	+544.6	+37.2	−0.6	73	+1008	+24	+1
24	+564	+36	0	74	+1012	+24	+1
25	+584	+34	0	75	+1017	+24	+1
26	+604	+33	0	76	+1021	+24	+1
27	+623	+32	0	77	+1026	+24	+1
28	+641	+31	0	78	+1030	+24	+1
29	+658	+30	0	79	+1034	+24	+1
30	+675.2	+28.5	−0.1	80	+1038.5	+23.7	+0.9
31	+691	+27	0	81	+1043	+24	+1
32	+706	+26	0	82	+1047	+24	+1
33	+720	+26	0	83	+1051	+24	+1
34	+734	+25	0	84	+1055	+24	+1
35	+748	+24	0	85	+1059	+24	+1
36	+760	+24	0	86	+1062	+23	+1
37	+772	+23	0	87	+1066	+23	+1
38	+784	+23	0	88	+1069	+23	+1
39	+794	+22	0	89	+1072	+23	+1
40	+804.0	+22.0	+0.3	90	+1074.9	+23.0	+0.9
41	+814	+22	0	91	+1078	+23	+1
42	+822	+22	0	92	+1080	+23	+1
43	+831	+22	0	93	+1082	+22	+1
44	+839	+22	0	94	+1084	+22	+1
45	+847	+22	0	95	+1086	+22	+1
46	+855	+22	0	96	+1088	+21	+1
47	+862	+22	0	97	+1089	+21	+1
48	+870	+22	0	98	+1091	+20	+1
49	+876	+22	0	99	+1093	+20	+1
50	+883.1	+21.9	+0.5	100	+1094.3	+19.1	+0.8

PART 3

DENSITY OF ETHYL ALCOHOL AND OF ITS MIXTURES WITH WATER

By N. S. Osborne

Part 3 consists of the determination of the density at a single temperature, 25° C, of pure ethyl alcohol and of various mixtures of alcohol and water in accurately determined proportions and the derivation of alcoholometric tables.

VI. DESCRIPTION OF APPARATUS AND EXPERIMENTAL WORK

The fundamental determinations of the density of pure alcohol were made simultaneously with its preparation, as previously described. The compounding of the mixtures, using portions of the same purified material, and the determination of their densities followed these fundamental determinations.

The density determinations were made partly by the method of hydrostatic weighing, described in Part 2 of this paper (p. 371), and partly by the use of specially constructed picnometers. By the use of these picnometers the effect of dissolved air upon the density of alcohol and of its mixtures was investigated to ascertain its significance in alcoholometry.

1. THE PICNOMETERS

Three picnometers, specially designed for the purpose on the principle of the Ostwald-Sprengel type, were used. In the construction of these picnometers the elements desired were as follows:

(a) A form adapted to the rapid attainment of the constant temperature of a surrounding water bath.

(b) Means of filling with minimum contact of liquid with air.

(c) Protection after filling from change of weight by evaporation or absorption of moisture; and

(d) Precision of filling.

The form of the picnometers is illustrated in Figs. 10 and 11. They were all made of Jena 16^{III} glass and thoroughly annealed. They are adapted to immersion in the constant temperature bath



Fig. 10 (one-fourth size).

already described. One of the picnometers (No. 274, Fig. 11) was of the Rudolphi⁹⁵² form, consisting of a hollow cylinder, which permits a rapid attainment of temperature but at expense of total volume. The others (Nos. 275 and 276, Fig. 11) were of the plain cylindrical form. The cap with stopcock attached, as shown in Fig. 11, is used to control the internal pressure when filling the picnometer. The bulb containing the liquid to be investigated is joined to the picnometer by the ground joint *b*. By proper manipulation, such as inclining the picnometer at a suitable angle and properly varying the air pressure, liquid is introduced into the picnometer. With the picnometer in position in the water bath, leaving only the upper portions of the capillaries emergent, the adjustment of the quantity of liquid is approximated as the temperature approaches constancy. Liquid may be introduced if necessary by means of a pipette placed with its tip to the aperture of the capillary with proper adjustment of pressure through the tube *c*. Small quantities may be removed by means of a strip of filter paper applied at the tip of the capillary. Enough liquid is removed to bring the meniscus just to the line *e* on the other capillary. The inside of the enlargement *d* of the tube is dried, either by means of filter paper or by a stream of dry air.

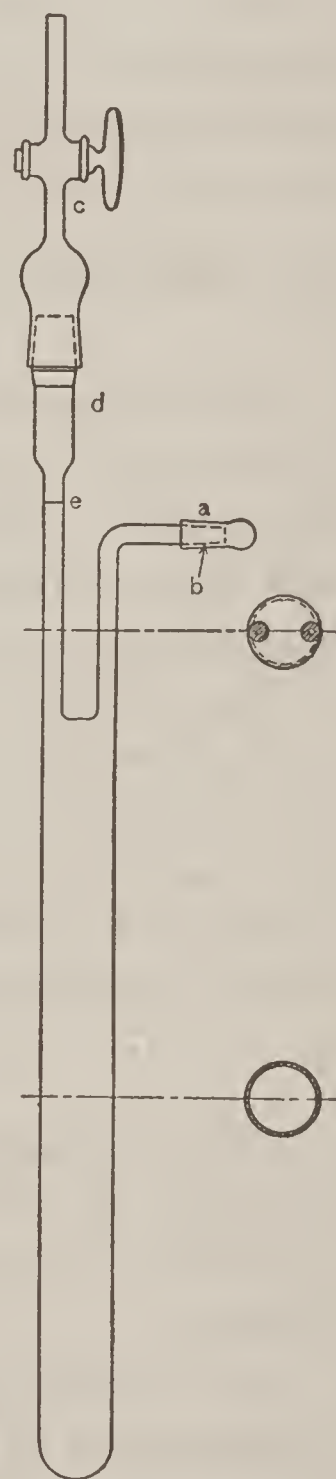


Fig. 11 (one-fourth size).

The picnometer itself serves as a sensitive thermo-indicator, and until the temperature becomes constant the final adjustment

of the quantity of liquid can not be made. When the temperature appears steady, 5 or 10 minutes more are allowed as margin of safety, and the filling is then completed. The picnometer is closed to prevent evaporation or absorption of moisture by means of the cap *f*, shown in the illustration.

The constants of these picnometers are given in Table XXXVIII.

TABLE XXXVIII
Constants of Picnometers

Picnometer B. S. No.	External volume at 25° C	Mass	Internal volume at 25° C
	ml	g	ml
274	92. 865	103. 1393	46. 1329
275	124. 56	75. 8659	87. 4046
276	123. 33	74. 4672	86. 0145

For the determinations of the volumes of these picnometers the density of water at $\frac{25^{\circ}}{4}$ C according to Chappuis ¹¹³² (0.997077) was used.

2. EFFECT OF DISSOLVED AIR

The effect upon the density of absolute alcohol caused by dissolved air was first determined by the method of hydrostatic weighing. The density of the alcohol was observed after long contact with dry air. The sample in the tube was then evacuated until no more bubbles were evolved. The density was then redetermined. Dry air was then passed into the bottom of the tube and bubbled through the alcohol and the density again determined. This was repeated on a second sample of alcohol. The results are shown in Table XXXIX.

TABLE XXXIX
Effect of Dissolved Air on Density of Alcohol
SAMPLE OF FRACTION NO. 3 I

Original density at $\frac{25^{\circ}}{4}$ C	Density at $\frac{25^{\circ}}{4}$ C after evacuating	Density at $\frac{25^{\circ}}{4}$ C after passing in air
0. 785094	0. 785160	0. 785134
FRACTION NO. 5 I		
0. 785041	0. 785102	0. 785061

Upon again saturating the alcohol with air the density did not return quite to its original value. This may be due to absorption of moisture. The above results indicate clearly that absorbed air decreases the density of absolute alcohol very appreciably. This is contrary to the experimental result obtained by Mendeléeff,^{207, 241} who, by taking air-free alcohol and shaking it with supposedly dry air, observed an increase in the density.

The above preliminary experiments were repeated, using the picnometers for determining the densities. In the method of hydrostatic weighing the alcohol was in contact with the air while the determination was being made, thus leaving doubt as to the completeness of the observed effect. Hence it was anticipated that more consistent and uniform results would be possible with the picnometer. The results are shown in Table XL.

TABLE XL
Effect of Dissolved Air on Density of Alcohol
SAMPLE OF ABSOLUTE ALCOHOL IN PICNOMETER NO. 274

	$D_{4}^{25^{\circ}}$	Change in density
After evacuating to remove air.....	0.785120	} 0.000065
After shaking with dry air.....	.785055	

SAMPLE NO. 5 I

Original sample.....	0.785080	} 0.000091
After evacuating.....	.785171	
After shaking with dry air.....	.785097	} .000096
After shaking again with dry air.....	.785081	
After reevacuating.....	.785177	} .000080
After shaking again with dry air.....	.785093	
After reevacuating.....	.785173	
Mean change.....		.000083

The above results demonstrate the reproducibility of the effect, although not indicating to what extent it is dependent on the temperature. The mean observed effect of dissolved air at 25° is a decrease in density of 0.000083.

The quantity of air absorbed by absolute alcohol was approximately determined by weighing, after evacuating to remove the air, a quantity of alcohol in a flask provided with a ground stopper, to which was attached a tube having two stopcocks with a bulb between. Dry air was admitted, shaken, and the flask again weighed. This was repeated until the weight became constant, indicating that no more air was absorbed. Ninety-three and fourteen hundredths (93.14) grams of alcohol absorbed .019 gram of air at about 20° C. This corresponds to about .13 cc of air for each cubic centimeter of alcohol.

The effect of dissolved air upon the density is so significant that the condition of alcohol as to whether air free or saturated with air must be specified if the density is used as a criterion of the completeness of dehydration.

Many experimenters neglect to state whether their alcohol is air free or air saturated. Mendeléeff^{207 241} states that the alcohol used by him was air free, and he used as a test for this condition the fact that when mixed with water no bubbles were given off. This does not seem to be a sufficiently definite test, since mixtures of alcohol and water may contain air, and from the low values obtained by Mendeléeff for the density of absolute alcohol and the method employed by him in obtaining it air free, it seems very doubtful if he removed more than a small part of the dissolved air.

The approximate effect of dissolved air upon the density of alcohol-water mixtures is derived later from the density determinations upon those mixtures.

3. DETERMINATION OF THE DENSITY OF ETHYL ALCOHOL

Having determined the magnitude of the effect of dissolved air upon the density, and having observed the greater uniformity in the density of the alcohol when saturated with air than when deprived of but left in contact with air, the density determinations to serve as criteria for comparison of different samples of supposedly pure alcohol were made in the air-saturated condition. The complete results of these determinations are given in Part I.

In this place are given only the actual determinations upon the samples, which were regarded as being the purest prepared and on

that account chosen to establish the value for the density of absolute alcohol.

These samples are designated by the number of the experiment in their preparation as described in detail in Part I of this paper. The complete record of observations and reductions to obtain the final mean value for the density of pure ethyl alcohol at $\frac{25}{4}^{\circ}\text{C}$ are given in Tables XLI to XLV.

TABLE XLI

Determinations of Density of Alcohol From Experiment IV Hydrostatic Weighing. Sinker No. 7

Date: Apr. 18, 1910. Record, vol. 646, p. 153

Corrected temperature		Mean corrected temperature t	Apparent weight of sinker in liquid	True weight of sinker in liquid	Volume of sinker at observed temperature	Density of liquid at observed temperature $D_{\frac{t}{4}}$	Density of liquid at 25°C $D_{\frac{25}{4}}$	Fraction No.
Green 2499	Tonnellot 4653							
25.008	25.010	25°010	62.5444	62.5358	47.71845	0.785088	0.785097	1
25.008	25.015		62.5443					
25.013	25.015	25°014	62.5459	62.5373	47.71845	.785057	.785069	2
25.008	25.020		62.5459					
25.013	25.015	25°014	62.5464	62.5378	47.71845	.785046	.785058	3
25.008	25.020		62.5464					
25.018	25.020	25°015	62.5466	62.5380	47.71846	.785042	.785055	4
25.008	25.015		62.5466					
25.013	25.020	25°014	62.5469	62.5382	47.71845	.785038	.785050	5
25.008	25.015		62.5468					
25.013	25.020	25°019	62.5468	62.5382	47.71846	.785038	.785054	6
25.018	25.025		62.5468					

Thermometer corrections: No. 2499, +0.008; No. 4653, +0.020. Observed air density: 0.00116 g/cm³. Buoyancy correction on weights: 0.0086 g. Mass of sinker: 99.9990 g.

TABLE XLII

Determinations of Density of Alcohol from Experiment VII. Hydrostatic Weighing: Sinker No. 7

Date: May 11, 1910. Record, vol. 646, p. 176

Corrected tempera- ture		Mean cor- rected tempera- ture t	Apparent weight of sinker in liquid	True weight of sinker in liquid	Volume of sinker at observed temperature	Density of liquid at observed temperature D_4^t	Density of liquid at 25° C D_4^{25}	Frac- tion No.
Green 2499	Tonnclot 4653							
25. 028	25. 030	25. 030	g 62. 5469	g 62. 5382	ml 47. 71847	0. 785038	0. 785064	1
25. 028	25. 035							
25. 008	25. 020	25. 015	62. 5465	62. 5378	47. 71846	. 785046	. 785059	2
25. 013	25. 020							
25. 013	25. 020	25. 016	62. 5465	62. 5378	47. 71846	. 785046	. 785060	3
25. 013	25. 020							

Thermometer corrections: No. 2499, +0.008; No. 4653, +0.020. Observed air density: 0.00116 g/cm³. Buoyancy correction on weights: 0.0087 g. Mass of sinker: 99.9990 g.

TABLE XLIII

Determinations of Density of Absolute Alcohol from Experiment XII

Hydrostatic Weighing: Sinker No. 7

Date: Apr. 7, 11, 12, 1910. Record, vol. 646, p. 147

Corrected temperature		Mean corrected temperature t	Apparent weight of sinker in liquid	True weight of sinker in liquid	Volume of sinker at observed temperature	Density of liquid at observed temperature $D_{\frac{t}{4}}$	Density of liquid at 25° C $D_{\frac{25}{4}}$	Fraction No.
Green 2499	Tonnellot 4653							
25.008	25.010	25.012	^g 62.5465	^g 62.5378	^{ml} 47.71845	0.785046	0.785056	1
25.008	25.020		62.5466					
25.008	25.020	25.018	62.5464	62.5377	47.71846	.785048	.785063	^c 1
25.023	25.020		62.5466					
25.013	25.015	25.016	62.5463	62.5375	47.71846	.785053	.785067	2
25.018	25.020		62.5463					
25.013	25.015	25.018	62.5469	62.5382	47.71846	.785038	.785053	3
25.018	25.025		62.5471					
24.998	25.005	25.005	62.5464	62.5377	47.71844	.785049	.785053	4
25.003	25.015		62.5465					
25.013	25.020	25.015	62.5463	62.5376	47.71846	.785050	.785063	5
25.008	25.020		62.5465					
24.993	25.000	24.996	62.5460	62.5372	47.7144	.785059	.785056	6
24.993	25.000		62.5459					

^c After passing dry air through.

Thermometer corrections: No. 2499, +0.008; No. 4653, +0.020. Observed air density: 0.00118 g/cm³. Buoyancy correction on weights: 0.0088 g. Mass of sinker: 99.9990 g.

TABLE XLIV
Determinations of Density of Absolute Alcohol From Middle Fraction of Experiment VI

Record of Observations and Calculations. Record: Vol. 646, p. 166

Deter- mined in picnometer B. S. No.	Date	Corrected temperature		Mean corrected tempera- ture t	Apparent weight of picnometer in air	Air density Correction for air buoyancy	True weight of picnometer	Mass of con- tained liquid	Volume of con- tained liquid	Density of liquid at observed tem- perature D_4^t	Density of liquid at 25° D_4^{25}
		Green 2499	Tonnellot 4653								
276	May 3, 1910	25.008	25.020	25.014	g 144.3581	0.001159 .1245	g 144.4826	g 68.6167	ml 87.4046	0.785047	0.785059
		25.008	25.020								
274	May 4, 1910	24.998	25.005	25.002	139.2664	.001177 .0898	139.3562	36.2169	46.1329	.785056	.785058
		24.998	25.005								

Thermometer corrections: No. 2499, +0.008; No. 4653, +0.020.

TABLE XLV
The Density at $\frac{25^{\circ}}{4}$ of Pure Ethyl Alcohol Saturated with Air. Assembled
Results of Most Satisfactory Experiments

Experiment number	Fraction number	Method of determination	Density at $\frac{25^{\circ}}{4}$ $D_{\frac{25}{4}}$	Observed density— mean density $\times 10^6$
IV.....	3	Hydrostatic weighing...	0. 785058	0
	4	do.....	. 785055	−3
	5	do.....	. 785050	−8
	6	do.....	. 785054	−4
VI.....	2	Picnometer No. 276.....	. 785059	+1
	2	Picnometer No. 274.....	. 785058	0
VII.....	1	Hydrostatic weighing...	. 785064	+6
	2	do.....	. 785059	+1
	3	do.....	. 785060	+2
XII.....	1	do.....	. 785063	+5
	2	do.....	. 785067	+9
	3	do.....	. 785053	−5
	4	do.....	. 785053	−5
	5	do.....	. 785063	+5
	6	do.....	. 785056	−2
Mean.....			. 785058	

Comparative examination of the results of the density determinations furnishes an index of the order of magnitude of the accidental errors. In Table XLV the average deviation from the mean of the 15 determinations is about 0.000004. The error of the mean due to accidental sources is within 0.000001.

The absolute accuracy of the densities is limited by the precision with which the thermometers have served to reproduce the International Hydrogen Scale. Two thermometers that were compared with the primary standards of the Bureau were always used. The corrected temperature indications of these thermometers rarely differed by as much as $0^{\circ}.01$. However, in consequence of the uncertainties inherent in the ice-point determinations, the effect of sticking of the meniscus, of small errors in reading, and the assumptions concerning the properties of *verre dur* glass that are necessarily made in the reproduction of the International Hydrogen Scale, the order of accuracy that has been attained in the reproduction of this temperature scale is, as nearly as can be estimated, about $0^{\circ}.01$ C, corresponding to about 0.000009 in density of pure alcohol at 25° C. Hence an accuracy of 1 unit of the fifth decimal place for this constant is all that can be expected.

As the result of these experiments 0.78506 is taken as the density at 25° C in grams per milliliter of pure ethyl alcohol saturated with air at ordinary atmospheric pressure.

4. PREPARATION OF THE MIXTURES—APPARATUS AND METHOD USED

In order to render the preparation of the mixtures as free as possible from causes of error in determining the proportions of alcohol and water, the apparatus shown in Fig. 12 was employed. It has been shown (p. 408) that pure alcohol at 25° C absorbs about 0.02 per cent of its own weight of air, which diminishes the density about 0.00008. Marek ⁶¹⁴ has shown that the absorption of air does not appreciably change the density of water at 25° C.

When alcohol and water both saturated with air are mixed, air is given off. This constitutes a source of error in the composition which can not exceed 0.02 per cent and is doubtless considerably less. This error may, however, be avoided by using water and alcohol deprived of air. Another possible source of error when the alcohol and water used in mixtures are weighed at atmospheric pressure is the difficulty in ascertaining the density of the mixture of air and vapor in the vessel above the liquid. To avoid the two errors mentioned above and to enable also an approximate determination of the effect of dissolved air upon the mixtures of alcohol

and water to be made, the mixing apparatus was constructed to permit the weighing of the liquids when evacuated to their vapor pressure. Referring to Fig. 12, it is seen that the apparatus consists of three parts—a bulb *B* with one opening, a three-way stopcock *D*, and a funnel-shaped vessel *A*, all provided with ground joints to permit their being joined together. In use the ground joints and the stopcock were lubricated sufficiently to render

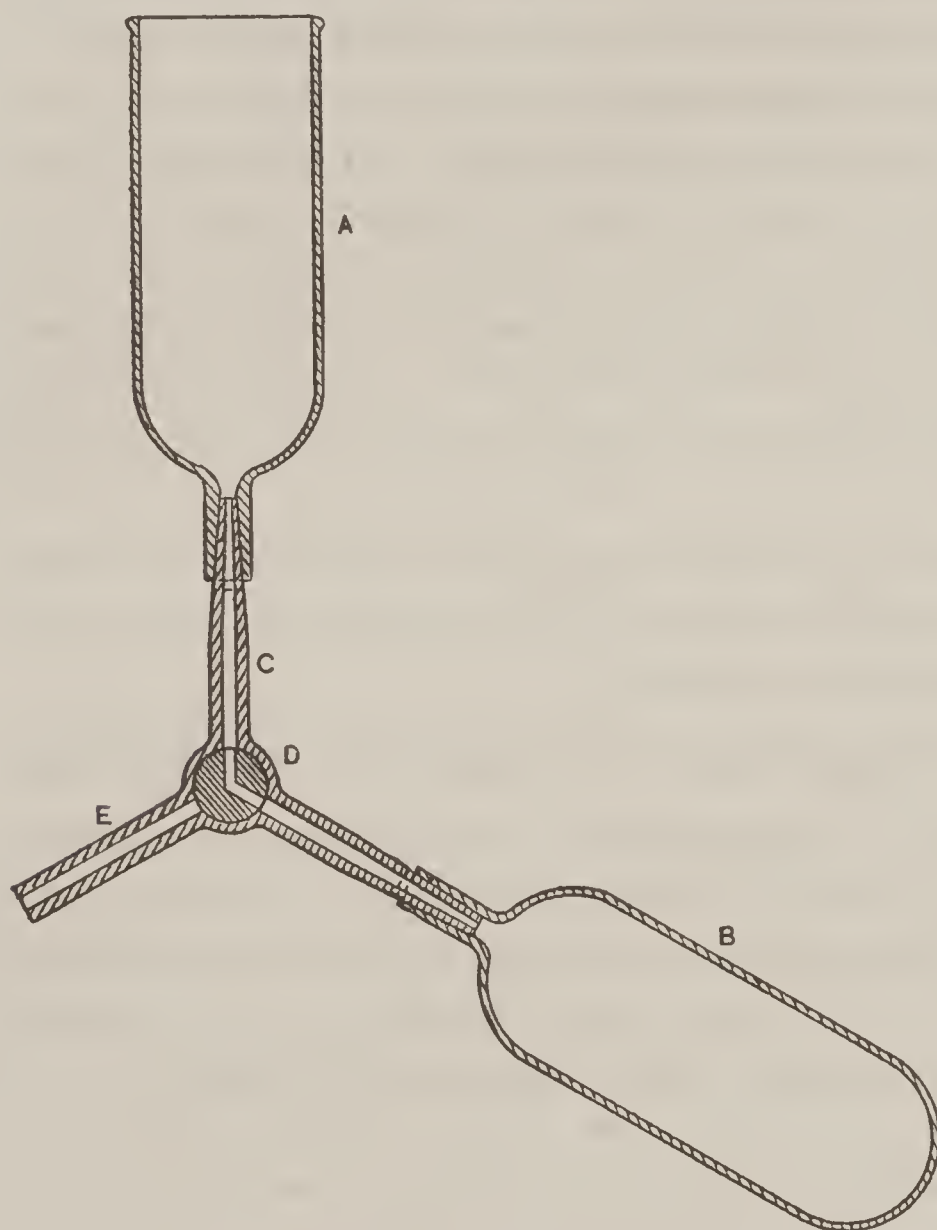


Fig. 12.—Mixing apparatus (one-third size).

them air tight, using a rather soft mixture of rubber and vaseline. The procedure in making a mixture was as follows:

The bulb with the three-way stopcock attached was first exhausted to remove the air and then closed and weighed. Next dry air was admitted and the stopcock removed. A suitable quantity of absolute alcohol was transferred to the bulb by means of a burette, only air which had been

dried by P_2O_5 being allowed to come in contact with the alcohol. The stopcock was replaced and the bulb again evacuated to remove the dissolved air from the alcohol. By repeated exhaustion and thorough shaking of the bulb the alcohol was freed from air. The stopcock was turned so as to close the passage to the bulb and connect the two other passages *C* and *E* which were then thoroughly dried by forcing through a current of dry air. The closed bulb containing only air-free alcohol and alcohol vapor was then weighed.

The water used in making the mixtures was freed from air by subjecting it in a flask to repeated exhaustion to a low pressure and shaking until no more air could be removed. The amount of water required to make a mixture of the desired concentration was calculated from the known quantity of alcohol already in the bulb. The tubular opening in the stopcock was first filled with water just to the bottom of the funnel and then closed. The calculated amount of water was then measured into the funnel by means of a burette. On turning the stopcock to connect the funnel with the bulb the outside atmospheric pressure forced the water into the exhausted bulb containing the alcohol. When the water again reached just to the bottom of the funnel the cock was turned so as to close the bulb and open the other passages. The funnel was removed and the cock again dried as before, after which the bulb containing the air-free mixture was weighed. Thus the relative proportions of alcohol and water were determined. Notwithstanding these precautions for excluding air from the mixtures a slight amount could still be detected in the form of bubbles evolved when the water was mixed with the alcohol at the very low pressure present, but it was certainly much less in amount than would be present by any method of mixing at atmospheric pressure. The mixtures may be considered practically air free. The possibility of the grease being dissolved by the mixture was investigated by evaporating 75 cc of a 95 per cent mixture. Only 0.2 mg of residue was found ($\frac{1}{300000}$ part).

The dimensions of this mixing bulb are as follows:

External volume, with stopcock.....cc..	209.7
Internal volume.....cc..	152
Mass with stopcock.....g..	134.9

The balance used in making the mixtures was No. 5485, a Rueprecht analytical balance of 600-g load, similar in design to that used for the density determinations and described on page 377. This balance was sensitive to 0.5 mg. Corrections for displaced air were applied to all weighings. The weights used in making the mixtures were set No. 5157, the same as those used for the density determinations and described on page 377, except that those from 500 mg to 10 mg were the ones belonging to the

balance. By previous test these were known to have no significant errors.

5. MATERIALS USED

The samples of absolute alcohol used in making the mixtures were portions of the distillates obtained in Experiments IV and XII, described in Part 1. The products of these experiments were regarded as of the greatest purity obtained.

The different samples of alcohol and the designations of the mixtures made from each, together with the determinations of density made at the time of use, are as follows:

SAMPLE 1

This sample was composed of fractions Nos. 2, 3, 4, and 5 from Experiment XII.

The density determinations were as follows:

Date	How made	$D_{4}^{25^{\circ}}$
1910		
Apr. 7.....	Hydrostatic weighing.....	0. 785058
Apr. 22.....	Picnometer No. 275.....	. 785055
Apr. 29.....	Picnometer No. 276.....	. 785043

From this sample were made mixtures designated as 95 D, 95 E, 95 F, 90 D, and 90 E.

SAMPLE 2

This sample was composed of fractions B, C, and D from Experiment IV.

The density determinations were as follows:

Date	How made	$D_{4}^{25^{\circ}}$
1910		
Apr. 18.....	Hydrostatic weighing.....	0. 785058
May 3.....	Picnometer No. 276.....	. 785053
May 6.....	Picnometer No. 274.....	. 785044
May 12.....	Picnometer No. 276.....	. 785055
Do.....	do.....	. 785047

From this sample were made mixtures designated 80 D, 70 D, 60 D, 50 D, 40 D, 65 D, 30 D, 20 D, 10 D, and 55 D.

SAMPLE 3

This sample was composed of fractions 2, 3, 4, 5, and 6 from Experiment IV. These portions are those upon which were made the original density determinations for Experiment IV. The density determinations were as follows:

Date	How made	D_{4}^{25}
1910		
Apr. 18.....	Hydrostatic weighing.....	0. 785057
May 25.....	Picnometer No. 276.....	. 785069
May 26.....do.....	. 785052

From this sample were made mixtures designated 75 D, 85 D, 45 D, 35 D.

SAMPLE 4

This sample was fraction A from Experiment IV. The density determinations were as follows:

Date	How made	D_{4}^{25}
1910		
Apr. 18.....	Hydrostatic weighing.....	0. 785084
July 5.....do.....	. 785088

From this sample were made mixtures designated 25 D, 15 D, 5 D, 2 D, 6 D, 99 D, and 98 D.

With the exception of sample 4, the original density determinations indicated these samples to be as free from water as any produced. The determinations at different dates show that within limits of experimental error no change in density had occurred. This was to be expected, since the portions were kept in bottles with well-ground stoppers, the bottles themselves being kept in a desiccator containing lime. In transferring the alcohol from one vessel to another air was always used which had been dried by phosphorous pentoxide, thus avoiding contamination by atmospheric moisture.

Sample 4 shows a density higher than that determined for the purest alcohol, and in calculating the percentage of the mixtures the percentage of alcohol in this sample is taken as 99.992, this figure corresponding to the density 0.785086.

6. EXPERIMENTAL WORK AND REDUCTION OF RESULTS

The record of the preparation of the mixtures is given in journal form in Table XLVI.

Several of the mixtures found necessary for interpolation purposes, of such proportions that the exclusion of air in their preparation was not considered essential, were made in an ordinary weighing flask at atmospheric pressure and without evacuating, as shown in the second part of Table XLVI. The record of all the density determinations made upon the mixtures is given in journal form in Table XLVII.

TABLE XLVI
Record of Preparation of Mixtures
AIR-FREE MIXTURES MADE IN SPECIAL MIXING BULB

Designation of mixture	Made from sample No.	Date when made	True weight of alcohol	True weight of mixture	Per cent alcohol in mixture
		1910	g	g	
95 D.....	1	Apr. 23....	66. 202	69. 820	94. 818
95 E.....	1	Apr. 25....	73. 570	77. 501	94. 928
95 F.....	1	Apr. 26....	101. 054	106. 414	94. 963
90 D.....	1	...do.....	90. 496	100. 543	90. 007
90 E.....	1	Apr. 30....	93. 542	103. 931	90. 004
80 D.....	2	May 4.....	79. 350	99. 254	79. 946
70 D.....	2	...do.....	79. 624	113. 679	70. 043
60 D.....	2	May 5.....	75. 695	126. 018	60. 067
50 D.....	2	May 6.....	61. 657	123. 319	49. 998
40 D.....	2	May 7.....	51. 249	126. 168	40. 651
65 D.....	2	May 9.....	79. 683	122. 586	65. 002
30 D.....	2	...do.....	39. 894	132. 968	30. 003
20 D.....	2	May 10.....	27. 077	135. 321	20. 009
10 D.....	2	May 11.....	13. 434	134. 231	10. 008
55 D.....	2	May 13.....	66. 701	121. 336	54. 972
75 D.....	3	May 25.....	66. 413	88. 592	74. 965
85 D.....	3	May 26.....	92. 576	108. 868	85. 035
45 D.....	3	May 27.....	58. 140	129. 345	44. 950
35 D.....	3	May 28.....	47. 300	134. 876	35. 069
25 D.....	4	June 1.....	34. 798	138. 692	<i>d</i> 25. 088
15 D.....	4	June 3.....	20. 243	134. 845	<i>d</i> 15. 010
5 D.....	4	...do.....	6. 984	139. 859	4. 994

MIXTURES MADE IN ORDINARY FLASK WITHOUT EXCLUDING AIR

		1910	g	g	
2 D.....	4	July 2....	2. 314	116. 407	1. 987
6 D.....	4	July 5....	6. 891	115. 888	5. 946
99 D.....	4	...do.....	91. 774	92. 732	<i>d</i> 98. 957
98 D.....	4	...do.....	90. 471	92. 310	<i>d</i> 97. 998

d Corrected for 0.01 per cent water in sample 4.

TABLE XLVII
Record of Determinations of Density of Mixtures
Assembled Results are Given in Table XLVIII

Designa- tion of mixture	Date of density de- termination	Condition of mixture	Method of determina- tion	Tempera- ture of determina- tion t	$D_{\frac{t}{4}}$	$D_{\frac{25}{4}}$
	1910					
95 D....	Apr. 23	Air free.....	Picnometer 275...	24. 988	0. 800486	0. 800476
95 D....	do.....	do.....	Picnometer 274...	25. 028	. 800440	. 800464
95 D....	do.....	Air saturated...	do.....	25. 016	. 800427	. 800441
95 D....	Apr. 25	do.....	do.....	24. 997	. 800431	. 800428
95 E....	do.....	Air free.....	Picnometer 275...	24. 990	. 800160	. 800151
95 F....	Apr. 26	do.....	do.....	24. 970	. 800077	. 800051
95 F....	do.....	do.....	do.....	24. 970	. 800087	. 800061
95 F....	do.....	Air saturated...	do.....	24. 976	. 800023	. 800004
95 F....	do.....	do.....	Hydrostatic.....	24. 979	. 800031	. 800013
95 F....	do.....	do.....	do.....	24. 978	. 800024	. 800005
95 F....	do.....	do.....	do.....	24. 976	. 800033	. 800012
95 F....	Apr. 27	do.....	do.....	24. 999	. 800018	. 800017
90 D....	do.....	Air free.....	Picnometer 275...	25. 003	. 813620	. 813623
90 D....	do.....	Air saturated...	do.....	24. 989	. 813608	. 813598
90 D....	Apr. 29	do.....	Hydrostatic.....	25. 008	. 813591	. 813598
90 E....	Apr. 30	Air free.....	Picnometer 276...	25. 008	. 813634	. 813641
90 E....	do.....	Air saturated...	do.....	25. 003	. 813614	. 813617
80 D....	May 4	Air free.....	do.....	24. 993	. 839297	. 839291
80 D....	do.....	Air saturated...	do.....	24. 993	. 839245	. 839239
80 D....	do.....	do.....	Hydrostatic.....	25. 008	. 839248	. 839255
70 D....	do.....	Air free.....	Picnometer 276...	24. 992	. 863344	. 863337
70 D....	May 5	Air saturated...	Hydrostatic.....	25. 008	. 863289	. 863296
60 D....	do.....	Air free.....	Picnometer 276...	25. 008	. 886856	. 886863
60 D....	do.....	Air saturated...	do.....	25. 000	. 886840	. 886840
60 D....	do.....	do.....	Picnometer 274...	25. 008	. 886820	. 886827
60 D....	May 6	do.....	Hydrostatic.....	25. 008	. 886831	. 886838
50 D....	do.....	Air free.....	Picnometer 276...	25. 003	. 909861	. 909863
50 D....	do.....	Air saturated...	do.....	25. 003	. 909859	. 909861
50 D....	May 7	do.....	Picnometer 274...	25. 006	. 909847	. 909852
40 D....	do.....	Air free.....	Picnometer 276...	24. 996	. 930140	. 930139
40 D....	do.....	Air saturated...	do.....	24. 994	. 930140	. 930135
65 D....	May 9	Air free.....	do.....	25. 016	. 875262	. 875276
65 D....	do.....	Air saturated...	do.....	25. 013	. 875253	. 875264
30 D....	May 10	Air free.....	do.....	25. 004	. 950682	. 950685
30 D....	do.....	Air saturated...	do.....	24. 994	. 950671	. 950667
20 D....	do.....	Air free.....	do.....	24. 983	. 966394	. 966393
20 D....	do.....	Air saturated...	do.....	24. 988	. 966380	. 966379
10 D....	May 12	Air free.....	do.....	25. 000	. 980431	. 980431
10 D....	do.....	Air saturated...	do.....	25. 000	. 980422	. 980422
55 D....	May 13	Air free.....	do.....	24. 990	. 898584	. 898576
55 D....	do.....	Air saturated...	do.....	24. 989	. 898575	. 898566
75 D....	May 25	Air free.....	do.....	24. 986	. 851456	. 851444
75 D....	do.....	Air saturated...	do.....	24. 970	. 851447	. 851421
85 D....	May 26	Air free.....	do.....	24. 994	. 826547	. 826542
85 D....	do.....	Air saturated...	do.....	24. 996	. 826511	. 826507

TABLE XLVII—Continued

Record of Determinations of Density of Mixtures—Continued

Designation of mixture	Date of density determination	Condition of mixture	Method of determination	Temperature of determination t	D_{4}^{t}	D_{4}^{25}
	1910					
45 D....	May 27	Air free.....	Picnometer 276 ..	24. 983	0. 920993	0. 920980
45 D....	...do....	Air saturated....do.....	24. 973	. 920980	. 920959
35 D....	May 28	Air free.....do.....	24. 980	. 941343	. 941329
35 D....	...do....	Air saturated....do.....	24. 986	. 941336	. 941326
25 D....	June 2	Air free.....do.....	24. 983	. 958810	. 958800
25 D....	...do....	Air saturated....do.....	24. 983	. 958820	. 958810
15 D....	June 3	Air free.....do.....	24. 986	. 973333	. 973328
15 D....	...do....	Air saturated....do.....	24. 980	. 973339	. 973331
5 D....	June 6	Air free.....do.....	24. 992	. 988171	. 988169
5 D....	...do....	Air saturated....do.....	24. 996	. 988177	. 988176
2 D....	July 2do.....	Hydrostatic.....	25. 070	. 993365	. 993383
6 D....	July 5do.....do.....	25. 045	. 986636	. 986648
99 D....	...do....do.....do.....	25. 046	. 788227	. 788267
98 D....	...do....do.....do.....	25. 042	. 791140	. 791176

The assembled results and their reduction to integral percentages are given in Table XLVIII. In this table also, columns 5 and 6, are given the observed and calculated values for the effect of dissolved air on the density.

The linear equation,

$$Y = 0.3326 X$$

in which *Y* represents the difference in density between air-free and air-saturated mixture, *X* the percentage of alcohol, and in which the coefficient is determined by a least square adjustment of the observed values of *Y* is used in calculating column 6.

It is seen that for mixtures of 95 per cent and under this equation represents the results within the limits of error of the determinations. If, however, the result for pure alcohol is considered, i. e., 0.000083, the equation is not satisfied within 0.00005. The results for this effect of air on the density of mixtures are given here not within any claim to their precision, but merely as a rough index of the possible effect. Further investigation by more exact methods would be of interest.

TABLE XLVIII

Determination of Density of Alcohol-Water Mixtures, Assembled Results and Reductions

Designation of mixture	Per cent alcohol by weight	Mean observed density at $\frac{25^{\circ}}{4}$		Air free $D_{\frac{25}{4}}$ minus air saturated $D_{\frac{25}{4}}$		Reduction of air saturated D_{25} to integral per cent	
		Air free	Air saturated	Obs $\times 10^6$	Cal $\times 10^6$	Per cent alcohol by weight	Density at $\frac{25^{\circ}}{4}$ C
2 D.....	1. 987	0. 993383	2	0. 993359
5 D.....	4. 994	0. 988169	. 988176	— 7	2	5	. 988166
6 D.....	5. 946 986648	6	. 986563
10 D.....	10. 008	. 980431	. 980422	9	3	10	. 980434
15 D.....	15. 010	. 973328	. 973331	— 3	5	15	. 973345
20 D.....	20. 009	. 966393	. 966379	14	7	20	. 966392
25 D.....	25. 088	. 958800	. 958810	—10	8	25	. 958946
30 D.....	30. 003	. 950685	. 950667	18	10	30	. 950672
35 D.....	35. 069	. 941329	. 941326	3	12	35	. 941459
40 D.....	40. 651	. 930139	. 930135	4	13	40	. 931483
45 D.....	44. 950	. 920980	. 920959	21	15	45	. 920850
50 D.....	49. 998	. 909863	. 909856	7	17	50	. 909852
55 D.....	54. 972	. 898576	. 898566	10	18	55	. 898502
60 D.....	60. 067	. 886863	. 886835	28	20	60	. 886990
65 D.....	65. 002	. 875276	. 875264	12	22	65	. 875269
70 D.....	70. 043	. 863337	. 863296	41	23	70	. 863399
75 D.....	74. 965	. 851444	. 851421	23	25	75	. 851336
80 D.....	79. 946	. 839291	. 839247	44	27	80	. 839114
85 D.....	85. 035	. 826542	. 826507	35	28	85	. 826596
90 D.....	90. 007	. 813623	. 813598	25	30	} 90	. 813622
90 E.....	90. 004	. 813641	. 813617	24	30		
95 D.....	94. 818	. 800470	. 800434	36	32	} 95	. 799912
95 F.....	94. 963	. 800056	. 800010	46	32		
98 D.....	97. 998 791176	98	. 791170
99 D.....	98. 957 788267	99	. 788135
100 (mean value).....	100. 000	. 785141	. 785058	83	33	100	. 785058

Table XLIX gives for every integral per cent the density at 10°, 15°, 20°, 25°, 30°, 35°, and 40° C in terms of the density of water at 4° C of mixtures of ethyl alcohol and water when saturated with air. The densities at 25° of integral per cents given in this table are derived from the experimental results of Table XLVIII by a method of interpolation partly graphical and partly analytical, the details of which are not of sufficient interest to be given here. The densities at 10°, 15°, 20°, 30°, 35°, and 40° are calculated from the densities at 25° by use of the equation

$$D_{\frac{t}{4}}^t = D_{\frac{25}{4}}^{25} + \alpha(t - 25) + \beta(t - 25)^2 + \gamma(t - 25)^3$$

The values of α , β , and γ are given in Table XXXVIIa. Page 402.

7. ALCOHOLOMETRIC DENSITY TABLE
TABLE XLIX

Density of Mixtures of Ethyl Alcohol and Water D_4^t

Per cent alcohol by weight	Temperature t						
	10° C	15° C	20° C	25° C	30° C	35° C	40° C
0	0. 99973	0. 99913	0. 99823	0. 99708	0. 99568	0. 99406	0. 99225
1	. 99785	. 99725	. 99636	. 99520	. 99379	. 99217	. 99034
2	. 99602	. 99542	. 99453	. 99336	. 99194	. 99031	. 98846
3	. 99426	. 99365	. 99275	. 99157	. 99014	. 98849	. 98663
4	. 99258	. 99195	. 99103	. 98984	. 98839	. 98672	. 98485
5	. 99098	. 99032	. 98938	. 98817	. 98670	. 98501	. 98311
6	. 98946	. 98877	. 98780	. 98656	. 98507	. 98335	. 98142
7	. 98801	. 98729	. 98627	. 98500	. 98347	. 98172	. 97975
8	. 98660	. 98584	. 98478	. 98346	. 98189	. 98009	. 97808
9	. 98524	. 98442	. 98331	. 98193	. 98031	. 97846	. 97641
10	. 98393	. 98304	. 98187	. 98043	. 97875	. 97685	. 97475
11	. 98267	. 98171	. 98047	. 97897	. 97723	. 97527	. 97312
12	. 98145	. 98041	. 97910	. 97753	. 97573	. 97371	. 97150
13	. 98026	. 97914	. 97775	. 97611	. 97424	. 97216	. 96989
14	. 97911	. 97790	. 97643	. 97472	. 97278	. 97063	. 96829
15	. 97800	. 97669	. 97514	. 97334	. 97133	. 96911	. 96670
16	. 97692	. 97552	. 97387	. 97199	. 96990	. 96760	. 96512
17	. 97583	. 97433	. 97259	. 97062	. 96844	. 96607	. 96352
18	. 97473	. 97313	. 97129	. 96923	. 96697	. 96452	. 96189
19	. 97363	. 97191	. 96997	. 96782	. 96547	. 96294	. 96023
20	. 97252	. 97068	. 96864	. 96639	. 96395	. 96134	. 95856
21	. 97139	. 96944	. 96729	. 96495	. 96242	. 95973	. 95687
22	. 97024	. 96818	. 96592	. 96348	. 96087	. 95809	. 95516
23	. 96907	. 96689	. 96453	. 96199	. 95929	. 95643	. 95343
24	. 96787	. 96558	. 96312	. 96048	. 95769	. 95476	. 95168
25	. 96665	. 96424	. 96168	. 95895	. 95607	. 95306	. 94991
26	. 96539	. 96287	. 96020	. 95738	. 95442	. 95133	. 94810
27	. 96406	. 96144	. 95867	. 95576	. 95272	. 94955	. 94625
28	. 96268	. 95996	. 95710	. 95410	. 95098	. 94774	. 94438
29	. 96125	. 95844	. 95548	. 95241	. 94922	. 94590	. 94248
30	. 95977	. 95686	. 95382	. 95067	. 94741	. 94403	. 94055
31	. 95823	. 95524	. 95212	. 94890	. 94557	. 94214	. 93860
32	. 95665	. 95357	. 95038	. 94709	. 94370	. 94021	. 93662
33	. 95502	. 95186	. 94860	. 94525	. 94180	. 93825	. 93461
34	. 95334	. 95011	. 94679	. 94337	. 93986	. 93626	. 93257
35	. 95162	. 94832	. 94494	. 94146	. 93790	. 93425	. 93051
36	. 94986	. 94650	. 94306	. 93952	. 93591	. 93221	. 92843
37	. 94805	. 94464	. 94114	. 93756	. 93390	. 93016	. 92634
38	. 94620	. 94273	. 93919	. 93556	. 93186	. 92808	. 92422
39	. 94431	. 94079	. 93720	. 93353	. 92979	. 92597	. 92208
40	. 94238	. 93882	. 93518	. 93148	. 92770	. 92385	. 91992
41	. 94042	. 93682	. 93314	. 92940	. 92558	. 92170	. 91774
42	. 93842	. 93478	. 93107	. 92729	. 92344	. 91952	. 91554
43	. 93639	. 93271	. 92897	. 92516	. 92128	. 91733	. 91332
44	. 93433	. 93062	. 92685	. 92301	. 91910	. 91513	. 91108
45	. 93226	. 92852	. 92472	. 92085	. 91692	. 91291	. 90884
46	. 93017	. 92640	. 92257	. 91868	. 91472	. 91069	. 90660
47	. 92806	. 92426	. 92041	. 91649	. 91250	. 90845	. 90434
48	. 92593	. 92211	. 91823	. 91429	. 91028	. 90621	. 90207
49	. 92379	. 91995	. 91604	. 91208	. 90805	. 90396	. 89979
50	. 92162	. 91776	. 91384	. 90985	. 90580	. 90168	. 89750

.TABLE XLIX—Continued

Per cent alcohol by weight	Temperature t						
	10° C	15° C	20° C	25° C	30° C	35° C	40° C
50	0. 92162	0. 91776	0. 91384	0. 90985	0. 90580	0. 90168	0. 89750
51	. 91943	. 91555	. 91160	. 90760	. 90353	. 89940	. 89519
52	. 91723	. 91333	. 90936	. 90534	. 90125	. 89710	. 89288
53	. 91502	. 91110	. 90711	. 90307	. 89896	. 89479	. 89056
54	. 91279	. 90885	. 90485	. 90079	. 89667	. 89248	. 88823
55	. 91055	. 90659	. 90258	. 89850	. 89437	. 89016	. 88589
56	. 90831	. 90433	. 90031	. 89621	. 89206	. 88784	. 88356
57	. 90607	. 90207	. 89803	. 89392	. 88975	. 88552	. 88122
58	. 90381	. 89980	. 89574	. 89162	. 88744	. 88319	. 87888
59	. 90154	. 89752	. 89344	. 88931	. 88512	. 88085	. 87650
60	. 89927	. 89523	. 89113	. 88699	. 88278	. 87851	. 87417
61	. 89698	. 89293	. 88882	. 88466	. 88044	. 87615	. 87180
62	. 89468	. 89062	. 88650	. 88233	. 87809	. 87379	. 86943
63	. 89237	. 88830	. 88417	. 87998	. 87574	. 87142	. 86705
64	. 89006	. 88597	. 88183	. 87763	. 87337	. 86905	. 86466
65	. 88774	. 88364	. 87948	. 87527	. 87100	. 86667	. 86227
66	. 88541	. 88130	. 87713	. 87291	. 86863	. 86429	. 85987
67	. 88308	. 87895	. 87477	. 87054	. 86625	. 86190	. 85747
68	. 88074	. 87660	. 87241	. 86817	. 86387	. 85950	. 85507
69	. 87839	. 87424	. 87004	. 86579	. 86148	. 85710	. 85266
70	. 87602	. 87187	. 86766	. 86340	. 85908	. 85470	. 85025
71	. 87365	. 86949	. 86527	. 86100	. 85667	. 85228	. 84783
72	. 87127	. 86710	. 86287	. 85859	. 85426	. 84986	. 84540
73	. 86888	. 86470	. 86047	. 85618	. 85184	. 84743	. 84297
74	. 86648	. 86229	. 85806	. 85376	. 84941	. 84500	. 84053
75	. 86408	. 85988	. 85564	. 85134	. 84698	. 84257	. 83809
76	. 86168	. 85747	. 85322	. 84891	. 84455	. 84013	. 83564
77	. 85927	. 85505	. 85079	. 84647	. 84211	. 83768	. 83319
78	. 85685	. 85262	. 84835	. 84403	. 83966	. 83523	. 83074
79	. 85442	. 85018	. 84590	. 84158	. 83720	. 83277	. 82827
80	. 85197	. 84772	. 84344	. 83911	. 83473	. 83029	. 82578
81	. 84950	. 84525	. 84096	. 83664	. 83224	. 82780	. 82329
82	. 84702	. 84277	. 83848	. 83415	. 82974	. 82530	. 82079
83	. 84453	. 84028	. 83599	. 83164	. 82724	. 82279	. 81828
84	. 84203	. 83777	. 83348	. 82913	. 82473	. 82027	. 81576
85	. 83951	. 83525	. 83095	. 82660	. 82220	. 81774	. 81322
86	. 83697	. 83271	. 82840	. 82405	. 81965	. 81519	. 81067
87	. 83441	. 83014	. 82583	. 82148	. 81708	. 81262	. 80811
88	. 83181	. 82754	. 82323	. 81888	. 81448	. 81003	. 80552
89	. 82919	. 82492	. 82062	. 81626	. 81186	. 80742	. 80291
90	. 82654	. 82227	. 81797	. 81362	. 80922	. 80478	. 80028
91	. 82386	. 81959	. 81529	. 81094	. 80655	. 80211	. 79761
92	. 82114	. 81688	. 81257	. 80823	. 80384	. 79941	. 79491
93	. 81839	. 81413	. 80983	. 80549	. 80111	. 79669	. 79220
94	. 81561	. 81134	. 80705	. 80272	. 79835	. 79393	. 78947
95	. 81278	. 80852	. 80424	. 79991	. 79555	. 79114	. 78670
96	. 80991	. 80566	. 80138	. 79706	. 79271	. 78831	. 78388
97	. 80698	. 80274	. 79846	. 79415	. 78981	. 78542	. 78100
98	. 80399	. 79975	. 79547	. 79117	. 78684	. 78247	. 77806
99	. 80094	. 79670	. 79243	. 78814	. 78382	. 77946	. 77507
100	. 79784	. 79360	. 78934	. 78506	. 78075	. 77641	. 77203

TABLE L
Comparison of Densities of Various Per Cents Alcohol by Weight

Per cent alcohol by weight	Bureau of Standards ¹³⁷⁶ 1910	Mendeléeff's values recalculated by—						Mende- léeff ^{207,241} 1865 ^f	B. S. 1910— Mend.	Kreit- ling ⁶⁴⁵ 1892 ^g	B. S. 1910— Kreit.
		Bureau of Stand- ards ¹²¹⁶ 1909	B. S. 1910— B. S. 1909	E. W. Mor- ley ¹⁰⁶⁰ 1904	B. S. 1910— Mor- ley	K. Normal- Eichungs- Komm. ⁶⁸⁵ 1893 ^e	B. S. 1910— K. N. E. K.				
	D ^{20°} _{4°} C	D ^{20°} _{4°} C	Units of fifth place	D ^{20°} _{4°} C	Units of fifth place	D ^{20°} _{4°} C	Units of fifth place	D ^{20°} _{4°} C	Units of fifth place	D ^{20°} _{4°} C	Units of fifth place
0	99823	99824	—1	99824	— 1	99824	— 1	99831	— 8	99823	0
5	98939	98936	+3	98927	+12	98948	— 9	98945	— 6	98941	— 2
10	98187	98185	+2	98189	— 2	98197	—10	98195	— 8	98194	— 7
15	97514	97522	—8	97519	— 5	97533	—19	97527	—13	97531	—17
20	96864	96870	—6	96872	— 8	96877	—13	96877	—13	96877	—13
25	96168	96171	—3	96175	— 7	96171	— 3	96185	—17	96175	— 7
30	95382	95385	—3	95387	— 5	95381	+ 1	95403	—21	95380	+ 2
35	94494	94499	—5	94506	—12	94496	— 2	94514	—20	94503	— 9
40	93519	93524	—5	93525	— 6	93523	— 4	93511	+ 8	93527	— 8
45	92472	92480	—8	92483	—11	92484	—12	92493	—21	92485	—13
50	91384	91386	—2	91386	— 2	91393	— 9	91400	—16	91392	— 8
55	90258	90262	—4	90263	— 5	90265	— 7	90275	—17	90263	— 5
60	89114	89115	—1	89115	— 1	89115	— 1	89129	—15	89114	0
65	87948	87950	—2	87950	— 2	87946	+ 2	87961	—13	87948	0
70	86766	86770	—4	86769	— 3	86762	+ 4	86781	—15	86767	— 1
75	85564	85570	—6	85569	— 5	85562	+ 2	85580	—16	85567	— 3
80	84344	84349	—5	84348	— 4	84339	+ 5	84366	—22	84347	— 3
85	83095	83097	—2	83097	— 2	83091	+ 4	83115	—20	83099	— 4
90	81797	81795	+2	81794	+ 3	81795	+ 2	81801	— 4	81801	— 4
95	80423	80417	+6	80421	+ 2	80424	— 1	80433	—10	80424	— 1
100	78934	78933	+1	78932	+ 2	78939	— 5	78945	—11	78930	+ 4

^e Recalculated from Mendeléeff's values of $D_{15^{\circ}\text{C}}^{20^{\circ}}$, given in Landolt and Börnstein (1905).

^f Mendeléeff's^{207,241} uncorrected values.

^g Recalculated from Kreitling's⁶⁴⁵ values of $D_{15^{\circ}\text{C}}^{20^{\circ}}$.

TABLE LI

Comparison of Densities of Various Per Cents Alcohol by Volume at 60° F
(15°56 C)

Per cent alcohol by volume at 60° F	Bureau of Standards ¹³⁷⁶ 1910	Mendeléeff's values recalculated by—				McCulloch ^{119h}	B. S. 1910—McC.	Squibb ^{433 i}	B. S. 1910—Squibb	Gilpin, ⁸ Drink-water, ¹⁰² and Squibb ^{433 i}	B. S. 1910—G., D., and S
		Bureau of Standards ¹²¹⁶ 1909	B. S. 1910—B. S. 1909	K. Normal-Eichungs Komm. ⁶⁴⁵	B. S. 1910—K. N. E. K.						
	D ^{60°} _{60°} F	D ^{60°} _{60°} F	Units of fifth place	D ^{60°} _{60°} F	Units of fifth place	D ^{60°} _{60°} F	Units of fifth place	D ^{60°} _{60°} F	Units of fifth place	D ^{60°} _{60°} F	Units of fifth place
0	1.00000	1.00000	0	1.00000	0	1.00000	0	1.0000	0	1.00000	0
5	.99282	.99283	— 1	.99279	+ 3	.99289	— 7	.9930	—18	.99281	+ 1
10	.98659	.98658	+ 1	.98657	+ 2	.98663	— 4	.9869	—31	.98660	— 1
15	.98104	.98112	— 8	.98114	—10	.98114	—10	.9815	—46	.98114	—10
20	.97595	.97607	—12	.97608	—13	.97600	— 5	.9760	— 5	.97608	—13
25	.97086	.97096	—10	.97097	—11	.97087	— 1	.9709	— 4	.97097	—11
30	.96535	.96540	— 5	.96541	— 6	.96541	— 6	.9652	+15	.96541	— 6
35	.95910	.95909	+ 1	.95910	0	.95915	— 5	.9593	—20	.95910	0
40	.95179	.95184	— 5	.95185	— 6	.95192	—13	.9519	—11	.95185	— 6
45	.94350	.94360	—10	.94364	—14	.94359	— 9	.9434	+10	.94364	—14
50	.93428	.93440	—12	.93445	—17	.93437	— 9	.9343	— 2	.93443	—15
55	.92419	.92435	—16	.92439	—20	.92427	— 8	.9242	— 1		
60	.91343	.91356	—13	.91358	—15	.91346	— 3	.9135	— 7		
65	.90202	.90210	— 8	.90214	—12	.90211	— 9	.9025	—48		
70	.88999	.89004	— 5	.89010	—11	.89003	— 4	.8900	— 1		
75	.87729	.87734	— 5	.87740	—11	.87730	— 1	.8769	+39		
80	.86381	.86389	— 8	.86395	—14	.86384	— 3	.8639	— 9		
85	.84942	.84955	—13	.84961	—19	.84950	— 8	.8496	—18		
90	.83381	.83394	—13	.83400	—19	.83385	— 4	.8340	—19		
95	.81604	.81611	— 7	.81616	—12	.81598	+ 6	.8164	—36		
100	.79388	.79387	+ 1	.79391	— 3	.79461	—73	.7946	—72		

^h From the table given in Gauger's Manual.^{215,1311.}ⁱ Bulletin No. 107 (Revised) Bureau of Chemistry.^{1265.}

8. REVIEW OF RESULTS

A comparison between the results of this investigation and those of other experimenters is shown in Tables L and LI. The results compared are the basis of several of the most important alcoholometric tables in general use at the present time. In order to render the different tables directly comparable, they have, as far as possible, been reduced to the same temperature basis.

In regard to the accuracy of Mendeléeff's results, reference to his publication ²⁴¹ of 1869 and examination of the tables of densities which he calculated by least square adjustment of the experimental data, shows differences between the observed and calculated results of several units of the fourth decimal place. Whether these discrepancies are attributed to experimental error or arise from defective adjustment, they furnish an indication of the magnitude of unexplained discordance. The adjusted values agree slightly better than the observed with the results presented here and have been used for comparison.

Explanation of the outstanding differences shown in the preceding comparison tables will not here be undertaken further than to suggest the possibility that a greater degree of refinement in apparatus and experimental conditions with regard to temperature control and measurement in particular have in the present work enabled the attainment of higher precision.

Taking into account all sources of error it would be difficult to account for any error in Table XLIX greater than .00004; however, the desirability of further experimental work to ascertain the accuracy with which the fundamental determinations can be repeated by another observer is suggested.

In conclusion, the author acknowledges indebtedness to Mr. H. W. Bearce for valuable assistance in the observations and calculations involved in the determinations.

WASHINGTON, August 1, 1910

PART 4

DENSITY OF ETHYL ALCOHOL AND OF ITS MIXTURES WITH WATER. (A CONFIRMATORY SERIES)

By H. W. Bearce

During December, 1910, and January, 1911, the work on ethyl alcohol and its mixtures with water was continued. A second and independent series of mixtures were prepared and their densities measured at 15° and 25° for the purpose of determining the reproducibility of the results already obtained. By making the determinations at both 15° and 25° it was possible to verify, at the same time, the densities at 25° and the rate of change of density between 15° and 25° .

VII. DESCRIPTION OF EXPERIMENTAL WORK

1. MATERIAL, APPARATUS, AND METHODS

The alcohol used for making the mixtures was from two new distillations of the same original material as that used in the first series.

A mixture of fractions 4, 6, and 8 of Experiment XIV (see p. 366, pt. 1) was used for making the mixtures designated in Table LIV as 10 E, 20 E, 30 G, 40 E, 60 E, 70 E, 80 E, and 90 E, while for making those mixtures designated as 20 G, 30 H, 50 E, and 70 F the mixed fractions for Experiment XV were used. The alcohol used for making the mixtures was in each case, immediately after distillation, placed in a glass receptacle closed by closely fitting ground joints. This receptacle was provided with a burette attachment through which the alcohol was drawn off as required for making the mixtures. Only air that had been passed through drying tubes containing calcium chloride and phosphorous pentoxide was allowed to come in contact with the alcohol.

The balances, weights, thermometers, sinker, and apparatus for controlling the temperature were those already described

All density determinations were made by the method of hydrostatic weighing. The methods of procedure that have been described in detail in parts 2 and 3 of this paper were, as far as possible, followed in part 4.

During this series of measurements ice-point readings of the thermometers were not taken after each temperature, but were taken at intervals sufficiently short to have shown any considerable change had it occurred. It is believed that the mean of the extended series of ice-point readings available from earlier work with the same thermometers, at the same temperatures, gives as consistent and as reliable results as could be obtained by individual readings.

For making the mixtures an ordinary graduated, stoppered flask was used instead of the special mixing bulb described in part 3. All mixtures were made with twice-distilled, air-free water, and to prevent expulsion of air bubbles at the higher temperature the mixtures were freed from excess of dissolved air by evacuating to about 1 cm of mercury and thoroughly shaking. At the time of determination they were probably in an approximately air-saturated condition.

In the following tabulated results no determinations have been omitted except those of two mixtures which were spoiled in the process of making and one other in which the temperature conditions were unsatisfactory. A comparison of these results with those given in part 3 of this paper is shown in Table LV, page 435.

2. EXPERIMENTAL RESULTS
TABLE LII
Determinations of Density of Alcohol from Experiments XIV and XV

Date	Experiment number	Fraction number	Corrected temperature		Mean corrected temperature t	Apparent weight of sinker in liquid	Air density within balance case	Air buoyancy on weights	True weight of sinker in liquid	Volume of sinker at observed temperature	Density of liquid at observed temperature $D_{\frac{t}{4}}$	Density of liquid at 15° $D_{\frac{15}{4}}$
			Tonnclot No. 4653	Haak No. 2040								
Dec. 9, 1910.	XIV	2	{ 15.158 15.178	{ 15.134 15.167	{ 15.159 }	62.1524	0.001190	0.0088	62.1436	47.70770	0.793486	0.793621
Do	XIV	4	{ 15.003 14.998	{ 14.992 14.992	{ 14.996 }	62.1465	.001190	.0088	62.1377	47.70752	.793613	.793610
Dec.12, 1910.	XIV	6	{ 15.058 15.058	{ 15.054 15.057	{ 15.057 }	62.1487	.001188	.0088	62.1399	47.70759	.793566	.793614
Do	XIV	8	{ 15.058 15.058	{ 15.062 15.067	{ 15.061 }	62.1488	.001188	.0088	62.1400	47.70759	.793563	.793615
Dec.28, 1910.	XIV	4, 6, and 8 mixed.	{ 14.958 14.958	{ 14.944 14.952	{ 14.953 }	62.1448	.001174	.0087	62.1361	47.70747	.793647	.793607
Jan. 23, 1911.	XV	Mixed fractions...	{ 15.033 15.018	{ 15.034 15.032	{ 15.029 }	62.1491	.001203	.0089	62.1402	47.70755	.793560	.793585
Jan. 30, 1911.	XV	do	{ 15.093 15.088	{ 15.092 15.092	{ 15.091 }	62.1507	.001162	.0086	62.1421	47.70762	.793519	.793596
Do	XV	{ Same sample as preceding	{ 14.948 14.956	{ 14.947 14.952	{ 14.951 }	62.1452	.001162	.0086	62.1366	47.70747	.793637	.793595
Mean												<i>i.</i> 793603

i XIV 2 not included.
Thermometer corrections: No. 4653, +0.°058 C; No. 2040, -0.°008 C. Mass of sinker No. 7, 99.9990 g.

TABLE LIII
Density of Alcohol from Experiments XIV and XV

Date	Experiment number	Fraction number	Corrected temperature		Mean corrected temperature t	Apparent weight of sinker in liquid	Air density within balance case	Air buoyancy on weights	True weight of sinker in liquid	Volume of sinker at observed temperature	Density of liquid at observed temperature D_t	Density of liquid at 25° D_{25}
			Tonnclot No. 4653	Haak No. 2040								
Jan. 11, 1911.	XIV.....	4, 6, and 8 mixed.	{ 24.993 24.993 24.968 24.993	{ 24.990 24.990 24.992 24.995	{ 24.992 24.987	62.5465	0.001182	0.0088	62.5377	47.71843	0.785049	0.785042
Jan. 23, 1911.	XV.....	Mixed fractions..	{ 24.968 24.993	{ 24.992 24.995	{ 24.987	62.5468	.001203	.0090	62.5378	47.71843	.785047	.785036
Jan. 30, 1911.	XV.....do.....	{ 25.068 25.073	{ 25.085 25.085	{ 25.078	62.5495	.001162	.0087	62.5408	47.71853	.784982	.785049
Do.....	XV.....	{ Same sample as preceding	{ 25.028 25.030	{ 25.038 25.038	{ 25.034	62.5473	.001162	.0087	62.5386	47.71847	.785029	.785058
Mean.....												.785046

Thermometer corrections: No. 4653, +0.°008 C; No. 2040, -0.°010 C. Mass of sinker No. 7, 99.9990 g.

TABLE LIV
Determination of Densities of Alcohol-Water Mixtures

Designation of mixture	Per cent alcohol by weight	Corrected temperature		Mean corrected temperature t	Apparent weight of sinker in liquid	Air density within balance case	Air buoyancy on weights	Volume of sinker at observed temperature	Density of liquid at observed temperature $D_{\frac{t}{4}}$	Density reduced to integral per cent and integral temp. $D_{\frac{t'}{4}}$	Integral per cent	Integral temperature t'
		Tonnelot No. 4653	Haak No. 2040									
		$^{\circ}\text{C}$	$^{\circ}\text{C}$									$^{\circ}\text{C}$
10 E.....	10.072	{15.006 15.008 25.056 25.058	{15.000 15.000 25.065 25.065	{15.004 15.004 25.061 25.061	53.1123	0.001181	0.0075	47.70752	0.982954	0.983053	10	15
10 E.....	10.072	{15.006 15.006 24.998 24.998	{15.002 15.002 24.990 24.990	{15.003 15.003 24.994 24.994	53.2273	.001181	.0075	47.71851	.980316	.980442	10	25
20 E.....	20.191	{15.006 15.006 24.998 24.998	{14.997 15.002 24.990 24.990	{15.003 15.002 24.994 24.990	53.7074	.001205	.0077	47.70752	.970482	.970716	20	15
20 E.....	20.191	{14.923 14.928 25.028 25.033	{14.920 14.924 25.045 25.045	{14.924 14.924 25.038 25.038	53.9042	.001205	.0077	47.71843	.966136	.966407	20	25
20 G.....	20.061	{15.113 15.103 14.996 14.993	{15.107 15.092 15.002 15.002	{15.104 15.098 14.998 14.998	53.6996	.001171	.0075	47.70743	.970643	.970687	20	15
20 G.....	20.061	{25.028 25.033 15.113 15.103	{25.045 25.045 15.107 15.092	{25.038 25.038 15.104 15.104	53.8967	.001171	.0075	47.71848	.966288	.966394	20	25
30 G.....	30.244	{14.996 14.993 24.958 24.983	{15.002 15.002 24.985 24.990	{14.998 14.998 24.979 24.979	54.3773	.001179	.0076	47.70763	.956436	.956891	30	15
30 H.....	29.889	{14.993 14.996 15.008 15.006	{15.002 15.002 14.997 15.002	{14.998 14.998 15.003 15.003	54.3487	.001201	.0078	47.70752	.957042	.956863	30	15
30 H.....	29.889	{24.983 24.983 15.008 15.006	{24.990 24.990 14.997 15.002	{24.979 24.979 15.003 15.003	54.6324	.001201	.0079	47.71842	.950880	.950673	30	25
40 E.....	39.970	{15.006 15.006 24.908 24.908	{15.002 15.002 24.910 24.912	{15.003 15.003 24.910 24.910	55.2152	.001182	.0078	47.70752	.938879	.938822	40	15
40 E.....	39.970	{24.908 24.908 14.958 14.958	{24.910 24.912 14.972 14.972	{24.910 24.910 14.965 14.965	55.5529	.001182	.0078	47.71834	.931589	.931460	40	25
50 E.....	49.879	{14.958 14.958 25.053 25.053	{14.972 14.972 25.070 25.070	{14.965 14.965 25.062 25.062	56.2084	.001200	.0080	47.70748	.918066	.917773	50	15
50 E.....	49.879	{25.053 25.053 24.958 24.958	{25.070 25.070 14.972 14.972	{25.062 25.062 14.965 14.965	56.5797	.001200	.0081	47.71851	.910074	.909853	50	25

TABLE LIV—Continued

Designation of mixture	Per cent alcohol by weight	Corrected temperature		Mean corrected temperature t	Apparent weight of sinker in liquid	Air density within balance case	Air buoyancy on weights	Volume of sinker at observed temperature	Density of liquid at observed temperature D_4^t	Density reduced to integral per cent and integral temp. $D_4^{t'}$	Integral per cent	Integral temperature t'
		Tunnelot No. 4653	Haak No. 2040									
60 E.....	60. 197	{15. 038 15. 043	{15. 042 15. 042	}15. 041	57. 3207	. 001203	. 0082	47. 70756	. 894753	. 895239	60	° C 15
60 E.....	60. 197	{25. 033 25. 033	{25. 048 25. 050		57. 7052	. 001203	. 0083	47. 71848	. 886493	. 886985	60	25
70 E.....	70. 131	{15. 048 15. 053	{15. 047 15. 052	}15. 050	58. 4304	. 001164	. 0081	47. 70758	. 871490	. 871842	70	15
70 E.....	70. 131	{24. 908 24. 908	{24. 012 24. 012		58. 8196	. 001164	. 0081	47. 71834	. 863138	. 863374	70	25
70 F.....	69. 957	{15. 018 15. 013	{15. 017 15. 017	}15. 016	58. 4074	. 001165	. 0081	47. 70754	. 871973	. 871884	70	15
70 F.....	69. 957	{24. 908 24. 933	{24. 928 24. 940		58. 7984	. 001165	. 0082	47. 71836	. 863584	. 863418	70	25
80 E.....	80. 016	{15. 053	{15. 060	}15. 056	59. 5683	. 001171	. 0083	47. 70759	. 847643	. 847729	80	15
80 E.....	80. 016	{25. 028 25. 028	{25. 048 25. 045		59. 9703	. 001171	. 0084	47. 71848	. 839027	. 839099	80	25
90 E.....	89. 980	{14. 858 14. 858	{14. 877 14. 872	}14. 866	60. 7707	. 001182	. 0086	47. 70738	. 822449	. 822281	90	15
90 E.....	89. 980	{25. 008 25. 008	{25. 025 25. 025		61. 1818	. 001182	. 0086	47. 71846	. 813643	. 813604	90	25
100 (mean).....	100. 000 79360	100	15
100 (mean).....	100. 000 78505	100	25

Thermometer corrections: Tunnelot No. 4653, +0.058 C at 15°; +0.008 at 25° C; Haak No. 2040, −0.008 C at 15°; −0.010 at 25°. Mass of sinker No. 7, 99.9990 g.

TABLE LV
A Comparison of Results

Per cent alcohol by weight	$D_{4^{\circ}C}^{15^{\circ}}$ Osborne	$D_{4^{\circ}C}^{15^{\circ}}$ Bearce	(O-B) difference $\times 10^5$	$D_{4^{\circ}C}^{25^{\circ}}$ Osborne	$D_{4^{\circ}C}^{25^{\circ}}$ Bearce	(O-B) difference $\times 10^5$
0	0.99913	0.99913	0.99708	0.99708
10	.98304	.98305	-1	.98043	.98044	-1
20	.97069	.97070	-1	.96639	.96640	-1
30	.95686	.95688	-2	.95067	.95067	0
40	.93883	.93882	+1	.93148	.93146	+2
50	.91776	.91777	-1	.90985	.90985	0
60	.89523	.89524	-1	.88699	.88698	+1
70	.87187	.87186	+1	.86340	.86340	0
80	.84772	.84773	-1	.83911	.83910	+1
90	.82228	.82228	0	.81362	.81360	+2
100	.79360	.79360	0	.78506	.78505	+1

PART 5

BIBLIOGRAPHY

By E. C. McKelvy

VIII. BIBLIOGRAPHY OF THE LITERATURE ON ALCOHOL AND ALCOHOLOMETRY

This bibliography has been compiled to meet the need of workers in the field of alcoholometry and all those interested in the physical properties and purification of the alcohols. It is hoped that the field has been covered reasonably completely. Especial attention has been given to completeness with respect to the purification and physical properties of ethyl alcohol and the points discussed in the present paper. It is considered that practically every physical property has a possible application in alcoholometry and in the tests that go to show the existence of a chemically pure substance. The three factors most important in the choosing of a physical constant for determining the composition of alcohol-water mixtures, or any other binary mixture, are, first, sensitiveness, determined by the rate of change of the given constant with change in composition; second, accuracy of the determination of the physical constant; third, ease and range of applicability. The first, though depending somewhat on the second and third factors, is more or less fixed, being essentially dependent on the nature of the components. The second and third factors depend largely on the state of experimental physical science, and consequently change as science progresses. The study of the relation between physical constants and composition in the binary mixture ethyl alcohol-water, in order to get light on the first factor above mentioned, has been taken up thoroughly for only a few constants. The density and thermal expansion have been studied most extensively because of their early application to alcoholometry. In the determination of the physical constants of pure ethyl alcohol, as a rule, little attention has been paid to

the possible water content and its effect. A critical analysis of the results already obtained on other constants besides those above mentioned and the redetermination of many of them are contemplated at this Bureau. The compilation of this bibliography is a preliminary step in that direction.

No attempt has been made to cover the more technological parts of the subject or the chemistry of alcoholic fermentations. Acknowledgment is made of the valuable help given by Dr. W. Bein, of the "Kaiserliche Normal-Eichungs Kommission," Berlin, in the preparation of this part of the work.

1. REFERENCES IN CHRONOLOGICAL ORDER

Each reference is made up, where the complete information is available, of the name of author or authors, the journal in which presented, the extent of the article, and the year in which published except where the chronological arrangement shows that information. The references of special interest with respect to the work of this paper are provided with an asterisk. Special parts of extensive articles are indicated by the page number. Notices of the abstracts are also included in the majority of cases.

The references are arranged chronologically by years and alphabetically within the year. They are numbered consecutively. The accompanying numbers in italics refer to the decimal classification of the subject matter and show the subjects treated of in the article in a much more complete manner than the title.

The abbreviations are, with few exceptions, those used by Chemical Abstracts, published by the American Chemical Society. C. B. has been used to indicate "Chemisches Zentralblatt" and J. B. the "Jahresberichte über die Fortschritte der Chemie und verwandter Teile anderer Wissenschaften."

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